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AEROSOLS AND FISSION
PRODUCT TRANSPORT

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AEROSOLS AND FISSION PRODUCT TRANSPORTABSTRACT

A survey is presented of current knowledge of the possible role of aerosols in the consequences of in- and out-of-core LOCAs and of end fitting failures in CANDU reactors. An extensive literature search has been made of research on the behaviour of aerosols in possible accidents in water moderated and cooled reactors and the results of various studies compared.

It is recommended that further work should be undertaken on the formation of aerosols during these possible accidents and to study their subsequent behaviour. It is also recommended that the fission product behaviour computer code FISSCON II should be re-examined to determine whether it reflects the advances incorporated in other codes developed for light water reactors which have been extensively compared.

RÉSUMÉ

Le présent rapport présente un aperçu des connaissances actuelles sur le rôle possible des aérosols par rapport aux conséquences des pertes de caloporteur primaire à l'intérieur et à l'extérieur du cœur, ainsi que des défaillances des raccords d'extrémité dans les réacteurs CANDU. Une recherche détaillée dans la documentation relative au comportement des aérosols en cas d'accidents possibles dans les réacteurs modérés et refroidis à l'eau, et les résultats de diverses études ont été comparés.

On recommande d'entreprendre d'autres travaux sur la formation d'aérosols durant ces accidents possibles et d'étudier leur comportement ultérieur. On recommande également de réexaminer le code informatique FISSCON II sur le comportement des produits de fission afin de déterminer s'il comprend les améliorations apportées à d'autres codes qui ont été mis au point pour les réacteurs à eau légère et qui ont fait l'objet de comparaisons détaillées.

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1. INTRODUCTION

The possible environmental consequences of accidents to nuclear reactors can be studied in at least three ways:

- (i) from the experience of actual reactor accidents
- (ii) from the results of experiments designed to simulate the effects of accidents to reactors
- (iii) from theoretical studies

It is only common sense to rate the reliability of the conclusions from the three types of study in the order given. Yet this is not necessarily the correct ranking. Nuclear reactor accidents are so rare that it is most likely that the next accident will be entirely different from any that have gone before. In spite of the best intentions it is difficult to ensure that experimental conditions will be typical of any future accident and it may well be that theoretical considerations, even if unconfirmed by experiment, are more directly applicable to future accidents than any past accident or even the most seemingly realistic experiment. On the other hand even the most plausible theoretical treatments may bear little relation to reality. It seems therefore that any study of nuclear safety should treat past accidents with reserve, current experiments with scepticism and theoretical treatments with some suspicion. What is probably required, even more than in most other fields of scientific endeavour, is an open mind.

In view of the rarity of accidents to reactors it is perhaps surprising that most of the countries involved in nuclear energy maintained sizeable research programmes on the consequences of possible accidents throughout the fifties, sixties and seventies. The TMI-2 accident in 1979 caused an increase in the pace of these programmes. Numerous studies have been done on the role of aerosols in reactor accidents and the effects of possible aerosols on their consequences. Experiments have been performed to try to identify the nature of aerosols and the chemical forms of the fission products which may be generated in reactor accidents and computer codes have been written to try to forecast the behaviour of such aerosols. There have been extensive comparisons of the performance of different codes on standardised accident simulations. By far the greater part of this information was obtained in support of Light Water Reactors. There is much less information directly pertaining to the CANDU type of reactor. The Safety Analyses by Ontario Hydro (Bruce B, Pickering Restart and Darlington) are meticulous in their analysis of the thermal-hydraulic aspects of accident conditions but treat the behaviour of aerosols only in rather general terms. The present report attempts to identify areas in which information on aerosols relevant to CANDU accident situations is

lacking and to suggest ways in which any deficiencies might be remedied. To this end an extensive literature survey has been performed. The list of sources consulted is given in Appendix A.

2. SOURCE TERM FOR POSTULATED ACCIDENTS TO CANDU REACTORS

The source term for postulated accidents to CANDU reactors is derived using the computer code CURIES II (Archinoff, 1983). To quote from the abstract, CURIES II is a Fortran-IV computer code which:

- estimates the steady state fractional distribution of fission products among the free, grain-boundary and grain-bound components of the total inventory of a fuel element. The distribution is calculated as a function of element power and nuclide half life; it is derived for 24 specific nuclides, for each fuel element in a CANDU fuel channel of specified power.
- estimates the steady-state total inventory of 24 specific fission product nuclides in each fuel element in a CANDU fuel channel of specified power.
- estimates the activity released from failed fuel in a CANDU channel as a function of time during a post-shutdown transient, using the calculated steady state results in conjunction with input fuel temperature transients and sheath failure criteria, and built-in transient release models.

In estimating the activity release from failed fuel CURIES II assumes:

- instantaneous release of the element's free inventory upon sheath failure.
- transient release from grains released instantaneously from failed elements.
- total instantaneous release of the grain boundary inventory when the fuel temperature reaches an input value.
- a portion of the grain-boundary inventory is released from failed fuel upon rewet; the portion released depends on the fuel temperature at rewet.
- an input fraction of the grain and grain-boundary inventory is instantaneously released when the fuel temperature reaches 1850 C. The postulated release mechanism is dissolution of uranium oxide by the Zircaloy sheath.

- following sheath failure, enhanced release from the uranium oxide grains can be accounted for via a model which considers enhanced grain growth when hot uranium oxide is exposed to steam for prolonged periods; release via this mechanism is added to any transient diffusional release from the grains.

Each of these mechanisms is considered in detail. It is beyond the scope of the present work to determine how well CURIUS II forecasts the actual releases of fission products from fuel under the specified conditions. Suffice it to say that it appears to be comprehensive and it is convincing.

2.1 IODINE

Because of its high volatility and its behaviour in the ecological chain (iodine - grass - cow - milk - babies - thyroid) iodine has generally been regarded as one of the radiologically most important isotopes likely to be released in a reactor accident. A very useful summary of significant reactor accidents has been given by Morewitz (1981) and is summarised in Appendix A2.1. In it he details the iodine releases from a series of accidents and it is apparent that the major iodine releases to the environment were from 'dry' accidents. For example while thirty times as many curies of the noble gases were released to atmosphere from TMI-2 as from Windscale, the iodine releases from TMI-2 were less than 0.1% of those at Windscale. A possible explanation was found in a paper by Adamson (1976) in a study of the behaviour of volatile reactive fission products in LMFBRs. He concluded from thermodynamic data and out of pile experiments that 'iodine will migrate exclusively as CsI(v) . Deposition of CsI(s) will occur at the cladding inner surface, but even at temperatures of 550 to 750 C its vapour pressure is sufficiently high for it to tend to migrate axially toward blanket and plenum zones.' Tellurium is also known to migrate down temperature gradients accompanied by caesium in oxide environments. Adamson reports that the major portion of fission product caesium released from hot fuel is transported down the radial temperature gradient as Cs(v) or, under certain conditions, as CsOH . According to Morewitz (1981) there is a chemical reason both for the limited release of iodine and the delay in releasing it when water is present. First the molecular form of iodine in the fuel is CsI , not elemental iodine, because there is ten times as much caesium produced in fission as iodine and, since both are volatile, they migrate to the cooler regions where they combine. CsI is one of the most stable compounds of iodine but it easily dissolves in water to form caesium (+ve) and iodine (-ve) ions. The only way to obtain gaseous forms of iodine (molecular iodine vapour, hypoiodous acid or methyl iodide) from a water solution of CsI is either to oxidise the ions slowly by contacting the water surface with air, or to form organic iodides by slowly reacting the ions with hydrocarbon materials (paint etc). If the solution is made basic (by the addition of NaOH or CsOH for

example) the oxidation of the iodine ion is largely prevented. Cubicciotti and Sanecki (1978), in a paper which predates the TMI-2 accident, found crystals of caesium iodide on the inside surface of power reactor cladding from a fuel rod which had exhibited PCI failure. In this 16 page paper (of which a synopsis is given in Appendix A2.2) the reference to caesium iodide is: "Some cubic crystals were observed in a linear deposit. They are shown in fig 4a. Analysis of one such crystal is shown in fig 4b in comparison with that of a chemically pure crystal of CsI". This is the only reference to caesium iodide in the paper and it is this sentence which is presumably referred to when the paper is quoted as the authority for the statement (paragraph 3.5.2.5.4.2 of the Bruce B Safety Report) "indeed caesium iodide (CsI) has been identified as the dominant species of iodine inside irradiated fuel".

There appears to be little other experimental evidence for the presence of CsI in fuel cans. Hobbins et al, 1986, (Appendix A2.3), describing work on the release of fission products from irradiated fuel as a function of temperature say "early results from the Argonne experiments have identified only the atomic species of iodine, caesium and tellurium at temperatures up to 1773K in fuel irradiated to a burnup of 31gWD/t" and "Raman and luminescence spectroscopy have been used at the Idaho National Laboratory to investigate the chemical forms deposited on surfaces in the effluent handling system downstream of the fuel in the PBF-SFD tests.... To date CsI has not been identified". In the STEP experiments, (Appendix A2.4), several accidents to LWRs were simulated in an in-pile loop and the fission products emitted from the overheated fuel studied. From the preliminary reports the experimental evidence for the presence of caesium iodide is somewhat equivocal as the following quotations indicate:

- "in low pressure high fuel heat up conditions the deposition samples consisted primarily of tin from the cladding together with caesium, molybdenum and rubidium. Iodine (often in this study associated with caesium) appeared less frequently".
- "Iodine and tellurium were not observed in samples from tests at low heat up rates".
- "Deposits containing molybdenum, caesium and iodine were generally amorphous, although some were definitely crystalline in nature".
- "Iodine is believed to combine with caesium in the fuel forming CsI, which is highly soluble in water. It is postulated that condensing steam would dissolve the CsI, also that CsI has a high dew point and would therefore condense to form aerosol particles in the cooler regions of the reactor, where they would be captured by deposition on surfaces during transport".

In STEP 1 (AD), which corresponded to a large coolant pipe break in a PWR with failure of emergency core cooling, rapid depressurisation and prompt core uncovering, "iodine was not detected on the upper or lower coupons. During an initial examination iodine with caesium was observed on the middle coupon in the elongated structures on splatters, but could not be located again when the energy spectra were being recorded".

In STEP 2 (corresponding to a transient event in a BWR combined with failures of high pressure emergency core cooling and long term decay heat removal. Automatic depressurisation and temporary low pressure coolant injection results in a very long time to core uncovering), "smooth rods found ... on the lower two coupons contained caesium, iodine and silver ... The ratio of the Cs/I peaks is consistent with that for CsI for all the deposits except for the long caesium, iodine and silver deposits on the upper coupon where the iodine peak is higher than for CsI".

In STEP 3 (TMLB), (a PWR transient with failure of main and auxiliary feedwater and failure to recover electric power; no depressurisation, coolant loss by venting through relief valves, core uncovering in one hour or more), "Agglomerated structures containing silicon, caesium, iron, tin, molybdenum and copper were present. No iodine, tellurium, antimony or uranium was detected".

In STEP 4, (similar to STEP 3 except that control rod simulant was included), "caesium was not identified and tin was not so prominent as in other tests. Iodine, tellurium, antimony and uranium were not found".

The above STEP results are quoted from EPRI NP-4967, Interim Report dated November 1986. An earlier report (Herceg et al, 1986) surprisingly gives rather more information on the STEP results on aerosol analysis than the EPRI Interim Report and says "Two distinct particle populations appear to be present, one of sub micrometre size and the other generally larger than a micrometre and often several tens of micrometres in at least one dimension. The smaller size particles outnumber the larger by many orders of magnitude. They appear to be formed by nucleation from the vapour phase and contain primarily Cs but also Sn, Rb, Mo and occasionally Te. Cs and I are found together, often with no other constituents and sometimes with Ag (presumably from deposition sample coupons) in the larger particles, usually having an elongated and crystalline form. Iodine is found only in association with Cs on all of the samples examined to date. Although molecular CsI has not yet been unequivocally identified, all available evidence points in this direction".

The conventional explanation for the presence of CsI in reactor fuel is that although the iodine and caesium atoms are formed at random in the fuel matrix, are surrounded by an overwhelming number of fuel molecules and therefore must be relatively far

apart from each other, they diffuse down the temperature gradient in the fuel element and meet one another at grain boundaries where they combine. It is often said that, since there are ten times as many fission product caesium atoms as iodine, caesium iodide must be the most likely form of iodine in the fuel.

If this is true it must be true for all reactors, including gas cooled, graphite moderated reactors such as Windscale and water cooled graphite moderated reactors such as Chernobyl. In the Windscale accident it is estimated (Chamberlain, 1981) that the ratio of Cs/I atoms released to the environment was about 70 about the same ratio as in the fuel. Chamberlain (1987) states that the ratios of I131 to Cs137 on air filters in the Windscale stacks and on an air sample taken in Sweden after the Chernobyl accident were the same ($I(\text{Bq})/Cs(\text{Bq}) = 20$, or $Cs(\text{atoms})/I(\text{atoms}) = 70$). There thus seem to be some similarities between the fission products released in the Windscale and Chernobyl accidents, even though one was gas cooled and the other water cooled.

None of this is really prejudicial to the formation of CsI within the fuel can, if not within the fuel itself. Garisto et al, 1986, (Appendix A2.5) point out that, on release in the form of CsI during the initial stages of fuel failure, the reducing steam conditions expected in the primary heat transport system (due to high pressure and hydrogen production by the zirconium steam reaction) should prevent fuel oxidation. Fuel oxidation could lead to the release of molecular iodine due to the entrapment of caesium in the fuel by the formation of caesium uranates. Although the RBMK 1000 reactors at Chernobyl were water cooled it is probable that by the time the cans had melted off and the fuel bared there was no water left and that air had access to both the fuel and the graphite moderator. Graphite and uranium oxide would be competing for oxygen but Windscale has shown that fuel oxidation and iodine release can occur in these conditions. While the operating temperature of the graphite was much higher at Chernobyl than at Windscale, the Windscale graphite heated up rapidly during the accident.

Garisto et al conclude that the thermodynamic stability of CsI, compared to all other species that can be formed in the PHTS, ensures that, for $Cs/I > 1$, CsI is the predominant species under all the conditions (pressure, temperature, Cs/I ratio, hydrogen/water ratio) studied. On the other hand in a hydrogen/steam atmosphere, in the absence of caesium, HI is the predominant iodine species at all temperatures.

Both CsI and HI dissociate in contact with water to form the involatile iodine ionic species. Therefore if CsI and HI are formed in the PHTS and contact water they will reach the containment vessel in an involatile form. Kinetic calculations show that for temperatures greater than 1000 K, the dominant iodine species will reach a steady state in less than 0.1s. The worst case is an accident where all the caesium is released and reacts to form CsOH before the release of atomic iodine. Even

under these conditions CsI becomes the dominant iodine species in less than 0.1s.

Should CsI not contact water the evidence is (Clough, 1983) that it will be deposited out quickly. Clough heated CsI and Te (in two separate experiments) to 700 C in a 200 cm/s flow of argon. The vapours of both species deposited out quickly in the cooler regions of the furnace tube (from 600 to 200 C), maximum deposition occurring in the 500 C region of the furnace tube. No mention is made of any aerosol formation in these experiments. Similar experiments by May (1958) in which sodium iodide was heated at 700 C in a stream of air showed that no particulate aerosol was formed even though 50% of the sodium iodide evaporated from the furnace and condensed in the cooler regions of the tube instead of being oxidised to elemental iodine.

The author was fortunate to have the benefit of a discussion with Dr J. Paquette of Whiteshell Nuclear Research Establishment, AECL where a great deal of significant iodine work has been done (Paquette et al, 1985, Paquette and Ford, 1985, Garisto et al, 1986 are some examples) and put forward the hypothesis that all existing results (including Windscale, Chernobyl and TMI) could be explained if it was accepted that iodine and caesium existed independently within the reactor fuel and never came close enough to combine chemically. In a Windscale type accident the two fission products were emitted independently, but in a TMI accident, in spite of the very low solubility of elemental iodine in water (0.029 and 0.078 g iodine per 100 ml of cold and hot water respectively) there was such a small mass of iodine and such a large mass of water available that the results could be explained by simple dissolution of elemental iodine in water. Dr Paquette argued convincingly that in that case the chemical results would be quite different from those actually found. It appears from the Whiteshell discussions that whatever the accident conditions when fission product iodine and caesium are released with water to the reactor containment, no release of airborne elemental iodine can occur provided the water in the containment is kept basic.

The origin of organic iodides at TMI-2 is uncertain. According to Garisto et al the PHTS is not a source. They tend to the view that reaction with organic impurities is a likely source. They believe that the most important airborne iodine during reactor accidents will be an organic iodide. Their conclusion is that insignificant quantities of airborne radioactive iodine would be released into the containment building. It is expected that organic iodide would form rapidly by reaction of any elemental and monatomic iodine with trace vapours in the containment atmosphere and that there would be sufficient organic contaminant in the containment atmosphere to convert the amounts of iodine likely to penetrate to the containment to organic iodide. (Barnes et al, 1967).

The only conditions envisaged in which elemental iodine can be directly injected into the containment are those involving end

fitting failures with fuel bundles ejected from the core. This will be accompanied by a considerable jet of coolant which will result in spray induced washout of the iodine. A jet spray washout model (Fluke et al, 1983) was developed for this case and when applied, for example, to the Pickering NGS case results in a washout factor of 19 for elemental iodine. In view of the fact that research work is still under way to determine the fraction of droplet carryover and iodine desorption a washout factor of only five is credited to jet washout in the Pickering Restart Analysis.

2.2 PARTICULATE AEROSOLS

The preliminary results of the STEP programme have been reported above. The statement that the sub micrometre particles outnumber the larger particles by many orders of magnitude implies also that the sub micrometre mass of particles is much larger than the mass of particles greater than one micrometre. In the LACE series of experiments (Appendix 2.6), artificially created aerosols (thought, nevertheless, to be representative of aerosols which might be created in an LWR accident) were generated and released in quasi reactor containment conditions. The tests used insoluble (MnO) and soluble (CsOH) aerosols. Their aerodynamic mass median diameter ranged from 1.4 to 4.3 micrometres and the suspended mass concentration from 3 to 30 g/cubic metre. Two of the tests (LA1 and LA3) focus on a containment bypass accident in a large dry PWR (called a V sequence in WASH-1400) and are considered relevant to aerosol deposition and resuspension in pipes under other severe accident conditions in PWRs, BWRs and CANDU. In the V sequence it is postulated that two check valves fail in the emergency core cooling pipeline to the reactor vessel. Following valve failures, overpressure in a connected low pressure pipe is assumed to cause it to rupture allowing primary system coolant water to discharge into an adjacent building. The loss of cooling water from this rupture eventually leads to core overheating and the release of radioactive aerosols and gases from the reactor into the pipeline. The pipeline is 63mm diameter, 27m long with five 90 degree bends, four horizontal sections and two vertical sections. In test LA1 virtually all of the aerosol material (>98%) was retained in the 63mm test pipe. The diameter of the test pipe was enlarged to 300mm dia before entering an 825 cubic metre building which was vented to the atmosphere through a scrubber. In test LA3 more than 70% of the injected aerosol, soluble or insoluble, was retained in the 63mm test pipe.

The objective of test LA2 was to simulate a postulated accident where a failure to isolate containment occurred. In addition to measuring the quantity and particle size spectrum of the leaked aerosol it was intended to determine the effect of leak path location i.e. whether at the top or bottom of the containment vessel. The leaks were 18mm dia orifices situated 16.5 and 4m above the containment vessel bottom. 98% of the injected aerosol mass was recovered and the fractions of settled, plated and

leaked aerosol mass were 0.501, 0.114 and 0.385 respectively. No significant difference was found in the mass of aerosol leaked through the upper and lower leak paths. This result is of some importance because most aerosol behaviour codes (see later) assume that the aerosol is well mixed and this is generally regarded as being a conservative assumption and predicts an airborne aerosol concentration which is high at any particular time. Razzaghi (1987) in a theoretical study of the vertical stratification of aerosols in a closed container, points out that the assumption of 'well mixed conservatism' is not necessarily true. About 64% of the aerosol entering the leak paths in test LA2 was deposited in the duct before entering the scrubber. This is a decontamination factor of 2.8 and shows that even a simple leak path (orifice plate in duct) can cause a significant reduction in the mass leaked.

No detailed results for test LA4 (a late containment failure) are given.

It seems unfortunate that, while the STEP experiments indicate that the aerosol likely to be formed in a reactor accident is predominantly sub micrometre, the LACE experiments used only aerosols greater than one micrometre diameter.

The DEMONA experiments, (Appendix A2.7), were designed to demonstrate the aerosol retention properties of a large dry PWR containment for core melt down accident sequences which involve late overpressure of the containment. The Batelle Frankfurt model containment was used (640 cubic metres) and equipped with steam and aerosol generators and appropriate thermal and aerosol instrumentation. Sub micrometre aerosols (tin oxide and silver) of primary particles size of 0.2 micrometre could be generated to give a maximum concentration of 12g/cubic metre. At the time of the report, six of the ten planned main experiments had been completed and the results show that aerosol depletion develops three or four times faster in a condensing steam-air atmosphere than in dry conditions, due to condensation and enhanced gravitational settling. At the end of the one hour aerosol injection period only about one third of the injected aerosol was still airborne and after one and a half hours the aerosol concentration had decreased by an order of magnitude. After 36 hours the concentration had decreased by a further 3 orders of magnitude. Only one per cent of the aerosol was plated out on walls and ceilings, confirming that the main removal process was sedimentation.

Even if the nature and particle size distribution of the aerosol formed in a reactor accident can be forecast with confidence, it is then necessary to predict its behaviour within the reactor and its containment and to estimate how much, if any, of the aerosol will escape to the environment. This is an exceedingly complex task which not only has to take account of the many aerosol removal mechanisms, but also the complex geometry of the reactor structures and the flow patterns which may occur in post accident conditions. Abbey and Dunbar in the second draft of Task VII of

the CSNI Task Force on Source Terms (undated and without references) (Appendix A2.8) review, mainly US, studies on fission product and aerosol transport in containment. They point out that modelling is complicated by the growth and evaporation of particles and mention that the removal rate, as a function of particle size, has a pronounced minimum between 0.2 and 2 micrometres diameter. They suggest that diffusion is relatively unimportant since, for dense aerosols, agglomeration is rapid and shifts the particle size upward, so that the removal rate is dominated by how fast particles can grow to a size at which gravitational settling becomes rapid. If the mean radii are between 0.1 and 1 micrometre and concentrations greater than 1g/cubic metre, agglomeration is an important mechanism of particle growth, although there is also a dip in the agglomeration rate between 0.2 and 2 micrometre diameter. For an initial size distribution near the most stable position it typically takes about 2 hours or less for a significant proportion of the particles to agglomerate to sizes where settling becomes rapid on the time scale of an hour. If conditions are such as to allow condensation of steam, growth is much faster but is very sensitive to thermal hydraulics.

Morewitz, 1982, (Appendix A2.9), considers the leakage of aerosols from containment buildings by reviewing experimental data on the plugging of ducts by aerosols and deriving a simple model. The model predicts that the mass of aerosol entering a duct before plugging is proportional to the cube of the duct diameter, the proportionality constant being a function of the effective density of the aerosol deposit and of the location of the plug. In the process of plugging leak paths, aerosols attach to the walls or to previously deposited aerosols. Some of these agglomerates break off and are resuspended in the airstream so that the sizes of aerosols escaping through short leak paths are substantially increased. When all of these effects are considered it appears that <1mg per day of respirable aerosol can leak from a typical reactor containment building operating at the design pressure and leak rate (0.1% vol/day). This is a reduction of more than four orders of magnitude from the case where the aerosol is assumed to leak as a gas.

3. COMPUTER CODES FOR FORECASTING AEROSOL BEHAVIOUR

Since 1975 a whole series of computer codes has been developed in an attempt to improve on the rather simple code CORRAL 2 used in the Reactor Safety Study, (Rasmussen, 1975) to describe the behaviour of aerosols released in reactor accidents. The following codes have been considered in this study viz:

- AEROSIM Appendix A3.1
- AEROSOLS Appendix A3.2
- HAA-4A Appendix A3.3

- NAUA-4 Appendix A3.4
- FISSCON II pp 14 to 15

Various comparison exercises between some of these codes and other codes have also been reviewed. These are summarised in the GREST code comparison exercise (Appendix A3.5), and the CEC Sensitivity Study (Appendix A3.6).

Butland et al, 1986 (Appendix A2.10), compare how the various codes treat different problems raised in two postulated PWR and two LWR accidents. They point out that the presence of hydrogen in certain accident sequences can reduce the density and viscosity of the carrier gas to such an extent as to modify aerosol behaviour significantly and to affect aerosol retention calculations by up to 30%. They state that most codes model four aerosol agglomeration mechanisms, Brownian, gravitational and two forms of turbulent agglomeration. The combined effect of the four mechanisms is calculated by adding the two turbulent and gravitational components in quadrature and then separately adding the Brownian component. It is not clear whether this is correct, but it may not matter since one component usually dominates. The main problem foreseen is that the formulae have not been verified for the high number concentration of particles predicted. Similarly a particle concentration of $5 \times 10^{13}/\text{m}^3$ is possible and the corresponding surface area of about $600\text{m}^2/\text{m}^3$ is much larger than the surface area of primary system structures so that chemical interaction between vapours and aerosols might be very important, e.g. an interaction between Te and Ag to form an aerosol would reduce tellurium retention in the primary system. Such reactions are not modelled in any of the primary system codes. Surface condensation of vapours on particles will also be important. In the accident scenarios referred to, condensation on particles outweighed condensation on surfaces by factors of 1.4 to 7, depending on the scenario.

It is also usual to model transport and retention by dividing the primary system into well mixed control volumes with unidirectional flow between them. New developments adopt a Lagrangian approach in which a plug of gas is followed through the primary system. This seems sensible for a pipe, but is less easy to justify for a large volume with possible recirculation.

The settling out of particles at $Re < 1$ is well described by the Stokes Law terminal velocity which is proportional to the square of the particle diameter and density and inversely proportional to the gas viscosity. For larger particles the settling velocity deviates from this law, but this doesn't really affect the issue since they settle out so quickly anyway. The ratio of the horizontal area of the vessel concerned to the volume is important in determining the rate of aerosol sedimentation. The effective settling area in any vessel with internal structures is not easy to determine because of possible flow channeling or recirculatory flows which could increase the effective areas. Other processes considered by Butland et al include turbulent

deposition (probably only important at high flow rates $>300\text{m/s}$ in fairly narrow and long pipes), diffusive deposition (only important for very small particles and probably only a minor contributor to deposition), thermophoretic deposition (also difficult to quantify in the absence of information on thermal gradients and boundary layer thicknesses), impaction and deposition on obstructions (these are not treated rigorously in existing codes, and better definition of flow fields and geometry is needed), resuspension and relocation, radioactive decay, radiolysis and photophoresis.

Schock et al, 1986, (Appendix 3.7) give a good description of the development of aerosol codes and the salient features of some of them. The CORRAL code (used in the Reactor Safety Study) assumed a single settling rate for the aerosols which was a universal function of the time since release and had only limited relevance to accident modelling. Modern codes either divide the aerosol size range into discrete size classes or make the more restrictive assumption that the size distribution is always log normal. In comparisons of codes it has been found that the discrete codes are in good agreement with each other and in better agreement with the experimental results than the log normal codes. The first aerosol code to incorporate steam condensation onto particles was NAUA (developed in the FRG). More recently, following experiments, it has had a diffusiophoresis model added to it. Two European fast reactor codes have recently been extended to take account of condensing steam - AEROSIM-M in the UK and AEROSOLS/B1 in France. In the USA similar developments led to the codes HAA-4, QUICK-M, and the MAEROS model in CONTAIN. and, in Japan, to the REMOVAL code. The major difference in the codes is in their modelling of condensation on the particles, especially where to make the boundary between the aerosol code and the thermal-hydraulic code from which it takes its input. This is discussed more fully by Butland et al. Once condensation has been modelled one has to distinguish between the water and non-water mass in the calculation. It is also useful to be able to follow radiologically important nuclides separately. One has to decide whether to allow the aerosol composition to be a function of particle size or to leave it independent. NAUA distinguishes between water and non-water mass as a function of size and can follow up to 50 nuclides on a size independent basis, while AEROSIM-M has size-dependent modelling of up to ten components, one of which can be water. MAEROS uses eight components.

Schock et al conclude that, subject to experimental validation and with the exception of the modelling of water condensation, the stand-alone aerosol codes are virtually in their final stage. On the other hand, no stand alone aerosol code is an exact solution to the condensation problem, since a scientifically correct model requires a coupled thermal-hydraulic and aerosol code. The CONTAIN code is such a fully coupled code but does not at present consider the effects of curvature and solutes on

condensation, although these could be incorporated.

3.1 GREST CODE COMPARISON

The GREST code comparison (Appendix A3.5) was a major test of a number of aerosol behaviour computer codes applied to specified accident conditions in LWRs viz:

1. AB hot leg sequence in Surry 1 PWR. A LOCA resulting from a large break in the primary system hot leg combined with a failure of AC power to the engineered safety features. Molten corium/concrete interactions following reactor vessel melt through were included but ex-vessel steam explosions were not.

2. S2CD cold leg sequence in Surry 1 PWR. LOCA due to a small break in the primary system cold leg, combined with the failure of the emergency core cooling injection system and containment spray system, but with the containment safety systems in operation. Extensive corium/concrete reactions and ex-vessel steam explosions were not considered.

3. Large hot leg break LOCA for the German Biblis B PWR. This assumes the reactor cavity was initially dry and that corium/concrete interaction took place immediately on pressure vessel melt through; no containment sprays; ECCS operates but conversion to recirculation core cooling fails, leading to core melting.

The results showed appreciable differences between the codes in all three problems, also differences in results from the same codes used by different organisations and between different versions of the same code. The log normal codes generally gave higher suspended mass concentrations and higher leaked mass than the discrete codes.

All of the codes generated similar (generally to 1%) values of the total retained mass (plated mass plus settled mass), but the partition between plated and settled fractions showed large variations (presumably due to different condensation and diffusiophoresis models used).

All of the codes preserve (or force) mass balance. However even the small mass unbalances are comparable to or larger than the total leaked mass.

Wet/dry effects vary considerably between the codes. In general HAA-4 showed the largest effects due to condensation on aerosol particles while NAUA-4 showed modest or negligible effects depending on who was using it. AEROSIM-2 showed small effects for Surry AB but larger effects for the other two problems.

In general the Biblis problem showed more scatter among the results than the Surry problems.

The log normal codes gave consistently higher values of the

accumulated leaked mass (the Source Term). Within a code group the scatter in the predicted leaked mass is only a factor of two. It is considered (Schock et al, 1986) that the calculational uncertainties in aerosol codes are probably small compared with those in other parts of the source term calculation.

3.2 FISSCON II

The discussion of FISSCON II, the Ontario Hydro fission product behaviour code has been left to the end so that it may be compared to codes which have been developed for LWRs. FISSCON II simulates the behaviour of fission products injected into the reactor containment for a given set of boundary conditions. It considers radioactive depletion and generation of fission products, their mass transfer between two major containment volumes, gravitational deposition, washout, leakage and a filtering system capable of recirculation and discharge. Fission product distribution in containment among the airborne and non-airborne phases and on the filter and outside of the containment can be described as a function of time and the release can be converted to radiation dose. The models used are judged unsuitable for short term transients and the code is used to calculate long term releases due to prolonged leakage or filtered air discharge.

In FISSCON II the generation of airborne activity is governed by:

- (a) radioactive pre-cursor decay
- (b) desorption from water or surfaces
- (c) desorption from filter
- (d) mass inflow from another volume
- (e) staged linear activity insertions to simulate delayed releases to containment

Depletion processes for airborne activity include:

- (a) radioactive decay
- (b) washout
- (c) plateout and chemisorption
- (d) filter removal
- (e) mass flow out of air volume

As described (Fluke and Jamieson, 1982) FISSCON II differs considerably from the codes already described. It deals exclusively with fission product vapours. Although the phrase 'fission product vapours and aerosols' is included in the introduction, there seems to be no reference to aerosols or the behaviour of particles in the equations used to assemble the code. Thus gravitational settlement, agglomeration of aerosols, diffusio-phoresis are not considered. Condensing steam is considered only to the extent that it may render surfaces wet and thus affect plateout. However, perhaps the weakest point is that there has apparently not been any comparison of FISSCON II with other codes in the manner described above. It can, of course, be argued that the code was derived for a unique reactor system and

there is therefore no point in comparing it with the others. That is true up to a point. However surely a fission product behaviour code should at least attempt to address the problem of particulate aerosols which will almost certainly be produced in any of the accident scenarios considered?

An important Canadian assessment of both the physical models used in the description of aerosol behaviour and of the numerical methods used in aerosol transport models is given by McDonald (1987,a,b). McDonald concentrates on the agglomeration of particles, their removal by deposition and their growth by condensation. He points out that in the development of models it has been necessary (a) to use models for spherical particles and to amend them by arbitrary shape factors; (b) to consider the physical effects as if each were independent of the other (c) to define arbitrary boundary layer thicknesses to account for physical gradients neglected in the basic analysis and (d) to make assumptions about saturation ratios in humid containment atmospheres. He states that in one set of computational tests the choice of Fuchs' or Pruppacher and Klett's collision efficiency for agglomerating particles appeared to make more difference to the total amount of aerosol released to the containment than did the inclusion of steam condensation and diffusio-phoresis, and concludes that there may be a measure of uncertainty in some of the very basic parameters.

In his discussion of numerical methods, McDonald (1987,(b)) examines the numerical methods most commonly used in the aerosol transport codes - the method of moments and discrete element models - as special cases of a general solution, the Method of Weighted Residuals. In RETAIN and HAA it is assumed that the particle size distribution is always log normal in which case there are three variables, the total particle number density, the geometric mean mass and the logarithm of the geometric standard deviation. The set of three ordinary differential equations is generated by integrating over the size dimension, with integrands weighted by the zeroth, first and second powers of mass, i.e. the first three moments. The integrations are performed analytically with agglomeration kernels and removal rates expanded as the first few terms of a power series. The method is efficient computationally but the restriction to a single shape for the size distribution and the need to express physical models as power series limit its practical application.

In the discrete element model (CONTAIN, REMOVAL, NAUA, AEROSIM) the size range is divided into a number of discrete elements or bins and it is assumed that particles exist only at the mid point sizes of the bins. Particle densities at these discrete sizes become the time dependent variables. An ordinary differential equation is generated for the time behaviour of each of these variables by integrating over each bin. The integrands are often weighted by mass and the integrations are performed numerically.

The method is flexible but it is essential to ensure that there are sufficient bins to accommodate the smallest and the largest particle and to provide enough detail in the distribution. In CONTAIN any particle which grows to a size larger than the largest expected particle is immediately put on the floor. This is a somewhat artificial device. Care must be taken that the numerical integration conserves mass.

McDonald concludes that there does not appear to be a single numerical method which is demonstrably superior to all the others for generating approximate solutions to the non linear integro differential equations describing aerosol particle behaviour. When used with skill and care each of the numerical methods should be able to produce reasonable answers to the mathematical problem. He stresses that some uncertainty still exists in the physical models used.

Work is proceeding at Whiteshell Nuclear Research Establishment with the objective of producing a computer model for calculating the generation, transport, deposition and release of fission product and structural material aerosols in the primary heat transport system of a CANDU reactor during severe fuel damage accidents. The approach is to use the Cathena code to calculate the thermalhydraulic conditions during accident scenarios and combine it with an aerosol code. The present (October 1987) status is that aerosol code models have been implemented and tested on the WNRE Vax and discussions have been initiated with Cathena development staff to determine the best way of combining aerosol code and Cathena models. (McDonald, 1987 (c)).

4. CANDU ACCIDENT SCENARIOS

4.1 Out of core loss of coolant accidents

This family of accidents calls into play all the major safety systems in the reactor - shutdown, emergency cooling and containment. According to Janzen (1981) the frequency of small diameter (less than 15cm) failures in nuclear quality piping is less than 1 per hundred reactor years and those in pipes greater than 15cm diameter, less than one in 10,000 reactor years. In the Safety Reports referred to above, the entire range of possible pipe failures is considered, up to the guillotine failure of the largest piping in the system - the reactor inlet and outlet headers. In any LOCA accident it is essential first that the reactor be shut down promptly, that emergency cooling is available and that any fission products released are contained so that individual and population doses are within the regulatory limits.

The analysis of LOCA accidents in the three safety reports studied (Bruce B, Pickering Start up and Darlington) deals with both small break (up to 700 kg/sec of coolant release) and large

break (greater than 1000 Kg/sec release). Breaks between 700 and 1000 kg per second release have some of the characteristics of small breaks and of large breaks and are treated separately.

The emergency coolant injection system is designed to ensure that for small LOCA scenarios there is no fuel or fuel channel damage. It is shown that there is no appreciable release of iodine 131 to the containment except when there is impairment of the Emergency Coolant Injection System (ECIS). The ultimate releases of iodine 131 to containment for various small break LOCAs are given in table 1 for Bruce B (taken from the Bruce B Safety Report Table 3.6.3-4)

TABLE 1

ULTIMATE RELEASES OF IODINE 131 TO CONTAINMENT FOR SMALL LOCAS

Accident scenario	Break Discharge Rate	Equilibrium Core % core inventory	Pre equilibrium core % core inv
LOCA ECI Avail	All	0.01	0.02
LOCA with loss of crash cooling	All	0.01	0.02
LOCA with loss of ECI, pumps working	950kg/s 150kg/s	22.7 4.7	25.8 6.6
LOCA with loss of ECI, pumps tripped	950kg/s 150kg/s	21.5 9.4	24.8 13.6
LOCA with loss of ECI Recovery Pumps	950kg/s 150kg/s	9.4 0.5	13.6 1.7
In Core LOCA with loss of ECI and CC (excluding releases from ejected fuel)	All	0.01	0.02
LOCA with loss of Class IV power	All	0.01	0.02

In the Darlington NGS A Safety Report, Appendix E, small LOCAs are defined as those with discharge rates of up to 1000kg/s. It is determined in the analysis that if ECI is available and forced flow circulation maintained, the fuel channels remain adequately cooled throughout the accident and activity releases are negligibly low. In accident scenarios in which a loss of forced circulation is postulated (i.e. LOCAs with loss of class IV power) the flow of coolant through the channels is maintained by thermosyphoning following rundown of the HT pumps. Should the

thermosyphoning cease in any channel, analysis shows that the maximum fuel temperature in a limiting channel could briefly peak at 770 C. No sheath strain failures are expected and any activity release would come from fuel elements with incipient sheath defects and would amount to much less than 5% of the core's free inventory of volatile fission products. Soluble and condensable species such as iodine and caesium would enter the containment as part of the liquid discharge. The only airborne species would be the noble gases. (E 2.5.1). In small LOCAs where the Emergency Coolant Injection System is not available the onset of fuel heat up and the time of the earliest sheath failures due to overheating are considerably delayed relative to the time of containment initiation. The containment pressure is sub atmospheric during the time when the fission products can enter the containment. The total release from the fuel is made up of two parts - the high temperature release during the early stages of the accident (HTR) and a post-accident release into the recirculating coolant as a result of leaching from the fuel matrix (LR). The quantities of the various fission products released for various times are given in table 2 (table E.2.5-1 of Darlington NGS A Safety Report)

Table 2

ULTIMATE LOECI RELEASE OF FISSION PRODUCTS FROM CORE TO
CONTAINMENT (% OF CORE INVENTORY) FOR STARVATION TIME OF 1015 SEC

Radionuclide	release at				
	HTR	HTR + LR	HTR + LR	HTR + LR	HTR + LR
	1015s	1d	10d	100d	365d
Kr 85m	19.0	19.1	20.4	26.3	26.4
Xe 135	13.9	14.1	15.4	21.3	21.4
Xe 133	19.9	20.0	21.4	27.2	27.4
Kr 85	17.6	17.8	19.1	24.9	25.1
Xe 131m	20.3	20.5	21.8	27.7	27.8
I 134	18.9	19.1	20.4	26.3	26.4
I 135	19.0	19.2	20.5	26.4	26.5
I 131	20.8	21.0	22.3	28.1	28.3
I 133	19.3	19.5	20.8	26.6	26.8
Cs 137	19.4	19.6	20.9	26.7	26.9
I 132	20.0	20.2	21.5	27.4	27.5
Sr 89	19.2	19.4	20.7	26.5	26.7

4.2 End fitting failures

This type of accident is of considerable interest for it is virtually the only conceivable accident to a CANDU reactor which results in the release of a fission product aerosol directly into the containment. End fitting failures are defined as failures of the fuel channel somewhere between the feeder pipe and the rolled joint. This would most probably cause a LOCA in the lower end of the small break spectrum. For the vast majority of cases the fuel bundles remain in the channel where they are well cooled by the flow of coolant to the break and fission product release would be negligible. The ejection of fuel from a channel can only result from highly improbable scenarios resulting from the complete loss of an end fitting. Even in this case the fuel bundle will probably be retained in the channel because of mechanical distortion of the channel circumference at the break. If the bundles escape from the channel they will most probably be caught near the reactor face by the surrounding feeder piping and be kept cool by discharge of coolant from the channel. Analyses of various possible modes of damage to fuel elements which may fall either to the fuelling machine duct or to the vault floor are made in the Darlington NGS A Safety Report section E 4.3 in which the results of some experiments in which 37 element bundles of unirradiated fuel were dropped in air a distance of 13.2 metres to a concrete floor covered with a 7mm thick carbon steel plate. In one test five elements failed and in the other two tests there were no failures. The two major possible mechanisms for fission product release from end fitting failures are first the burst of a sheath and the diffusional release of the free inventory and second the oxidation of the fuel. Both these mechanisms are considered in detail and estimates made of the probable release of fission products in both the short and long term. These releases are calculated in section E4.6 of Appendix E of the Darlington NGS A Safety report, table 4.3.3-1 of the Pickering Restart Safety Analysis, table 3.6.6.3-1 of the Bruce Safety report. The response of the containment system is considered with several postulated impairments, e.g. failure of isolation dampers to close on demand, deflated air lock seals, failure of auxiliary pressure relief valves to open on overpressure.

In calculating the quantities of fission products released, it is assumed that the fuel is ejected from a maximum power fuel channel and the coolant leak is 270kg/s. It is calculated that the sub atmospheric hold up period within the containment lasts for 22 hours before the start of a controlled release through the Filtered Air Discharge System (FADS). During this period the fission products decay and the atmospheric iodine content is limited by the equilibrium established between airborne iodine and that in the water. An effective partition coefficient of 10 is assumed (4.3.4.2 of Pickering NGS A Units 1 and 2 Restart analysis), implying that the atmospheric concentration of iodine is 0.01% of that in the water. It is assumed that this is all methyl iodide for which the efficiency of the FADS is 99.9%.

4.3 In core LOCA

The pressure tubes in a CANDU reactor form the primary pressure boundary of the high pressure coolant within the reactor core. The material (Z-2.5%Nb) and design are such as to show "leak before break" characteristics. A coolant leak would be detected by dew point monitoring in the annulus gas system. Leaks have been detected in this manner in the past, notably in Pickering NGS A, units 3 and 4, and in Bruce NGS 2 pressure tubes. One exception was the sudden rupture of the pressure tube in channel G-16 of Pickering NGS A (Zircaloy 2) which was shown to be due to a high concentration of radial hydrides. The unstable crack propagation and failure of the Zr-2.5 Nb pressure tube in channel N 06 of Bruce NGS Unit 2 in March 1986 occurred while the reactor was shut down, well after the leak was detected. Failure of the calandria tube in N 06 was attributed to the large water hammer loading associated with the highly sub cooled HT liquid at the time the pressure tube failed.

The scenario examined in the safety reports is the spontaneous failure of a pressure tube (pt) and its calandria tube (ct). This is a conservative treatment since, even in the unlikely event of a pressure tube rupture, the calandria tube is expected to remain intact. The maximum discharge which can result from a pt/ct guillotine failure is about 230kg/s and this maximum discharge is used to assess the possible damage to in-core structures. The response of the moderator system is analysed to determine if the rupture discs in the calandria relief pipe would burst, resulting in a discharge into the containment, causing a significant tritium release from the moderator. The assessment is performed assuming that neither the containment pressure nor the ventilation exhaust activity values reach levels which activate the containment isolation. This maximises the predicted dose for an in-core break.

Following a pt/ct failure, fuel may or may not be ejected into the moderator. For small longitudinal slits the fuel will remain in the channel and fuel damage will be minimal. For large longitudinal failures, ejection of several fuel bundles may occur. For a sudden circumferential rupture the two sections of the ruptured pressure tube may be misaligned and fuel bundles may be ejected as from the barrel of a gun.

Short term activity release from the ejected fuel may occur due to mechanical damage, by temporary and moderate fuel temperature excursions in the moderator, by fuel fragmentation on cooling and by leaching of fission products from the fuel fragments. The bulk of these releases, except for the noble gases, will be retained by the moderator. Activity release may continue in the long term by leaching of the affected fuel.

Airborne releases from the calandria vessel will be limited to tritium and the noble gases and other radiologically significant species will remain in the liquid. It seems therefore that the release of fission products from an in-core break is not an

important safety issue. The long term airborne concentration of fission products will be determined by the equilibrium partitioning between gaseous and liquid phases. In the Pickering Start Up Safety Analysis the source term derived for end fitting failure is also applied to the in-core LOCA and is conservative (4.4.7). See also Darlington Safety Report, Appendix E, 5.7 and Bruce B Safety Report 3.6.7

5. FISSION PRODUCT BEHAVIOUR IN THE CONTAINMENT

In the three safety reports studied, the most comprehensive account of the behaviour of aerosols in the containment is given in the Bruce B Safety Report (3.5.2.5.4.2). Tritium as vapour may be present in the containment as a result of heavy water discharged from either the heat transport system or the moderator. The concentration of tritiated heavy water can be calculated from the known water vapour pressure, assuming 100% relative humidity. The report classifies fission product aerosols as inert gases, stable vapours, unstable vapours and aerosols and their expected behaviour is summarised below:

Inert gases will exist mostly in the gaseous state since partitioning between air and water favours air. Charcoal in the FAD system will absorb some of them at least temporarily, but no credit is claimed for this in the analysis. The main removal mechanism is radioactive decay during the sub atmospheric hold up time, but radioactive noble gases will nevertheless dominate the atmospheric release.

Stable vapours such as some organic compounds of iodine, notably methyl iodide, are highly volatile and will remain airborne at low concentrations. Deposition of methyl iodide is slow, but it is removed (at 99.9% efficiency) by the FAD system. In the long term methyl iodide is expected to be the dominant airborne iodine species although the concentrations will be very low.

Unstable vapour such as elemental iodine can exist in the gaseous phase but deposits out rapidly onto containment surfaces or becomes dissolved in water. It is removed by the FAD system.

Aerosols will dominate the airborne radioactivity in the short term but are rapidly depleted by various natural processes including gravitational settlement, inertial impingement, Brownian diffusion, eddy diffusion, thermophoretic diffusion, diffusio-phoretic diffusion, all of which are size dependent.

6. CONCLUSIONS

The conclusions of this study into the effect of the presence of aerosols on specified CANDU reactor accidents are as follows:

1. The three safety documents studied - for Bruce B, Darlington and Pickering Restart are comprehensive, detailed and convincing in every respect except one - the treatment of aerosols which might arise in any of the specified accident sequences.

2. No attempt is made to characterise any aerosols which might arise. Ontario Hydro is a sponsor of the STEP and LACE studies but no use is made of the results, nor indeed is any mention made of them in the Safety Studies. The dates of Safety Documents are 1984, 1985 and 1986 while the STEP experiments started in 1982 and the LACE experiments were begun in 1985. It is surprising that there is not some reference to them at least in the later safety reports.

3 There is an extensive literature on aerosols arising from postulated reactor accidents. Most of it is directed specifically to LWRs, but much of the information is or could be applicable to CANDU. The authors of the safety reports do make use of the information on the release of iodine, but the effects of aerosols, except in the most general terms, are largely ignored. This may be because no conditions are envisaged in which aerosols can have a prominent role, but the safety reports negate this argument by stating (in relation to fission products in containment) "aerosols will dominate the airborne radioactivity in the short term but are rapidly depleted by various natural processes ...". However these processes are not quantified.

4. This is particularly so in the code FISSCON II which includes such processes as plateout and washout but not condensational growth due to condensation of steam on particles and subsequent accelerated gravitational settlement, diffusiophoresis, turbulent deposition etc. It may be argued that such effects would only increase the removal of aerosols from the reactor atmospheres and reduce subsequent releases to the environment which are already within permitted limits. That does not seem to be a valid argument for neglecting them. By comparison with the other codes referred to, FISSCON II is not a very sophisticated document.

5. To be fair, the LWR operators have the advantage of many independent groups working on similar problems and coming up with diverse solutions which they are able to compare at frequent meetings. Ontario Hydro and AECL attend such meetings, but must always be somewhat on the periphery of the discussion, except, of course, in the case of iodine behaviour. However the absence of opportunity for outside

comparison and criticism is in itself a reason for extra effort rather than less.

6. It is recommended that a reassessment should be made of the extent to which aerosols may arise in the specified CANDU reactor accident conditions especially in the light of the results from the STEP experiments. It is further recommended that a quantitative assessment be made of the extent to which such aerosols are removed in their path through the compartments of the reactor by the various mechanisms referred to above. It is suggested that it is insufficient to conclude that the penetration of a specified compartment will be below the recommended limits without doing a detailed assessment.

7. The computer code FISSCON II estimates the radiological dose incurred by people downwind of a release, but only from the airborne fission product cloud. It is also stated to be suitable only for calculating the effects of long term releases. No account is taken of deposition from the fission product cloud, presumably because it is estimated that any aerosol emitted during an accident will be below permitted levels. It is considered that realistic quantitative estimates should be made of such releases and the dose commitment due to their deposition, since it will very often be the deposited aerosol which will determine, in the event of a short term release, whether evacuation of the local population will be necessary and how long such an evacuation should last.

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APPENDIX A

A 1 LIST OF SOURCES SURVEYED

Papers with the identification IAEA-SM-281 are contained in the proceedings of the symposium "Source Term Evaluation for Accident Conditions" held at Columbus, Ohio, 28 October to 1 November 1985 and published by IAEA, Vienna, 1986).

F. Abbey and I.H. Dunbar CSNI Task Force on Source Terms - Task VII, Fission product/aerosol transport in Containment and other Buildings. 2nd Draft, undated

E.C. Beahm, W.E. Shockley and C.F. Weber Chemistry and transport of Iodine in Containment, IAEA-SM-281/41, 1986

A.T.D. Butland and M.R. Kuhlman Factors affecting primary system radionuclide retention in an LWR severe accident, IAEA-SM-281/20, 1986

D. Cubiciotti and J.E. Sanecki Characterisation of deposits on inside surfaces of LWR cladding, J Nuc Mat 78, pp96-111, 1978

R.J. Fluke and T.J. Jamieson FISSCON II: A fission product behaviour code, Ontario Hydro Report No 82004, Feb 1982

F. Garisto et al Radioiodine chemistry in reactor accident conditions, IAEA-SM-281/46, 1986

R.R. Hobbins et al In vessel release of radionuclides and generation of radionuclides, IAEA-SM-281/7, 1986

H. Jordan and V. Kogan Aerosol modelling in containment, IAEA-SM-281/45, 1986

T.F. Kanzleiter et al DEMONA experiments to investigate aerosol behaviour within a containment, IAEA-SM-281/43, 1986

LACE-LWR Aerosol containment experiments LACE TR-012, 1987

H.A. Morewitz Leakage of aerosols from containment buildings, Health Physics, 42, 2, Feb 1982

H.A. Morewitz Fission product and aerosol behaviour following degraded core accidents, Nuclear Tech, 53, pp 120-134, May 1981

H.A. Morewitz et al Results of the GREYST code comparison, CSNI Report No 116, OECD Nuclear Energy Agency, 1986

Nuclear Aerosols in Reactor Safety, SOAR, Nuclear Energy Agency, OECD, June 1979

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Nuclear Reactor Accident Source Terms, Nuclear Energy Agency.
OECD, 1986

The Source Term Experiments (STEP), Project Deposition Sample
Characterisation, EPRI NP-4967M, Nov 1986

W.O. Schock et al Behaviour and retention of fission products
in the containment, IAEA-SM-281/30, 1986

R.P. Wichner and R.D. Spence A chemical equilibrium estimate of
the aerosols produced in an overheated reactor core, Nuc Tech,
70, pp 376-393, Sept 1985

A2 SYNOPSIS OF SOURCES SURVEYED

A2.1 FISSION PRODUCT BEHAVIOUR AND AEROSOL BEHAVIOUR FOLLOWING DEGRADED CORE ACCIDENTS

by H.A. Morewitz Nuclear Technology, 53, pp 120-134, May 1981

ABSTRACT: A review of reactor accidents and destructive tests shows that when water was present, only a very small fraction of the volatile fission product inventory (other than the noble gases) was released to the environment. In addition the iodine component was released over a period of days. However in those situations where there was no water and the containment was poor, the release of volatile fission products was large and rapid.

The role of water in both limiting and delaying the release of iodine can be explained by the fact that the chemical form of iodine in intact fuel rods was caesium iodide (CsI). It is one of the most stable forms of iodine but it easily dissolves in water. The only ways to obtain gaseous forms of iodine from a water solution of CsI are by the slow processes of either contacting the solution surface with air or by reacting the solution with hydrocarbon materials (paint etc). Of the other volatile fission products, most dissolve in water. Rubidium, strontium, barium, caesium and their oxides dissolve in water after first reacting to form their respective hydroxides. Arsenic and selenium oxides are directly soluble in water. However tellurium and its compounds (except the alkali metal tellurides, the acid and the hydride) are largely insoluble in water. In reactor accidents and destruction tests where water was absent, up to 45% of the tellurium was released to the atmosphere in the form of fine particles (< 1µm in diameter), yet in all cases when water was present, no tellurium was released. This result may be due to the fallout of the fine particles due to growth caused by condensation of water vapour or to the solubility of Cs_2Te , since there is some evidence for this compound in intact fuel.

Vapourised fission products form aerosols as they condense. Analysis has shown that the aerodynamic sizes of agglomerated aerosols should increase as the concentration is increased, and this has been confirmed by a large number of experiments that indicate a size dependence with the cube root of concentration. Above about 30g/cubic metre concentration, the experiments show an even stronger dependence of aerosol size on concentration leading to the rapid formation of very large agglomerates. In addition when aerosols are released into a saturated steam atmosphere, the steam condenses on them and causes them to grow to still larger sizes. The large aerosols fall out in a short time so that the aerosol mass available for leakage is reduced. Furthermore in saturated steam atmospheres, leak paths are rapidly plugged with water so that the leakage of aerosols to the environment is dramatically restricted.

Due to the solubility of the volatile fission product compounds and the aerosol behaviour mechanisms, the offsite dispersion of radioactive materials (other than gases) following a major light water reactor accident will be small.

The paper gives a review of reactor accidents (table A.2.1) and destructive tests which indicate that the availability of water played a powerful role in both limiting the amount of fission products released and in delaying the release of radioiodine. In all LWR accidents and destructive tests, no more than 0.5% of the available iodine has ever been released to the atmosphere and only very limited quantities of the other fission products have been detected in the environment. The iodine has always been released over a period of days. However for those accidents and destructive tests where no water was present and the containment was poor (Windscale, KIWI-TNT, SNAPTRAN-2) up to 70% of the radioiodine was promptly released along with up to 67% of the other fission products.

TMI-2 ACCIDENT

In TMI-2 very little radioiodine was released to the environment (^{131}I) although 2.5 to 13 million Ci of krypton and xenon were released through the same path. The off-site noble gas release decreased exponentially with about a 12h half life so that most of the environmental release occurred in the first few days. The iodine release from the fuel handling and auxiliary building increased for a period of about 15 days before decreasing towards background. Obviously some chemical or physical mechanisms intervened to limit the total amount of radioiodine released to about 0.00001% while at the same time releasing around 5% of the Xe^{133} (figure A.1). Other mechanisms delayed the release of the iodine for days. No Cs^{137} or Sr^{90} was detected in the environment other than that due to fallout from weapons tests.

SL-1 ACCIDENT

In the SL-1 accident a very large fraction of the core was destroyed and most of the primary water was ejected from the primary system. The reactor building filled with steam which leaked to the environment from gaps in the doors on the operating floor, from open doors in the control room and from the exhaust on the fan floor. The expelled primary system water was highly contaminated with fission products, including iodine, as evident from examination of water saturated clothing of the victims.

Despite the fact that the reactor building was of ordinary construction and was not designed as a containment building, less than 0.5% of the I^{131} inventory was released to the atmosphere. During the first 16 hours after the accident only about 10 Ci of I^{131} was released to the environment and about 70 Ci over the next 30 days (figure A.2). Radioactive material found within the 3 acre exclusion area surrounding the building included about 0.1 Ci of Sr^{90} and about 0.5 Ci of Cs^{137} , but much of this material

TABLE A.2.1 SUMMARY OF REACTOR ACCIDENTS
from Morewitz, 1981

Facility	Dry/Wet	Contained Yes/No	Iodine	Fission Products	Noble Gases	Distance to Background (Miles)	Contam- inated thermal area	MW	MWd	Offsite manrem	Whole body dose	Thyroid dose
Windscale Dry		No	27,000 Ci to atm (12%)	1600 Ci Te 600 Ci Cs 137 80 Ci Sr 89 9 Ci Sr 90	540,000 Ci to atm	200+ (est)	200 sq miles(est)	250	4000	a	4.5 rem (site) <20 mrem (offsite)	9.5rem (adult) 16rem (child)
SL-1	Wet	To some extent	80 Ci to atm	0.1 Ci Sr 90 0.5 Ci Cs 137	10,000 Ci to atm	50	75 acres	3	932	0	3 fatalities 27 rem on site	5.5 rad on site .035 rad
NRX	Wet	Yes	a	10,000 Ci in 1,000,000 gal water in contain- ment	a	0.25	reactor building	30	a	0	16.1 rem on site	0 off site
TMI-2	Wet	Yes	17 Ci to atm	Not detected in atmosphere	10 MCi to atm	20	reactor building	2720	242,000	2,000	4.2 rem on site; 70 area off site	0 off site
WTR	Wet	Yes	0 Ci to atm	10,000 Ci in 1,600,000 gal water in contain- ment	<800 Ci to atm	1	"			0	2.1 rem on site; 0 off	0 off site
CR-3	Wet	Yes	70 Ci in 40,000 gal water; 2Ci in contain- ment air	a	10,000 Ci in contain- ment air	0	"	2452		0	0	0
PRTR	Wet	Yes	205 Ci (27%) in cont water 7 Ci (0.9%) in cont air		about 50% in cont air	>6	a	0.12	0.001	0	0	0
HTRE-3	Dry	No	34 Ci (14%) to atmosphere	0.1Ci Sr91 400Ci total to atmosphere	a	a	reactor building	24	1.66	a	a	a
ORR	Wet	Yes	0.15 to 2Ci to atmosphere	1000Ci in primary 300Ci l	a	a	reactor building slight	40	491	0	a	a
MTR	Wet	Yes	a	15 times normal in primary	a	a	reactor building minor	90	a	a	a	a
ETR	Wet	Yes	a	6.4 Ci to atm; 42Ci to pond	a							

TABLE A.2.1. (continued)

Facility	Dry Wet	Contained Yes/No	Iodine	Fission Products	Noble Gases	Distance to Contam Background Area (miles)	MW thermal	MWd	Offsite dose man rem	Whole body dose	Thyroid dose
NRU	Wet	Yes	a	Large release of to coolant and building; small release to environment	a	a	100 acres 200	a	a	19 rem on site	a

Notes: a not available

DESTRUCTIVE TESTS

Facility	Dry Wet	Contained Yes/No	Iodine	Fission Product	Noble Gas	Atmospheric Release	Dist to Bkgd miles	Contam area acres	Energy MWs
NRX Natural Uranium, Heavy Water Moderated, Research Reactor, Chalk River, Ontario									
TMI-2 Three Mile Island, Unit 2, Middletown, Pennsylvania									
WTR Westinghouse Test Reactor, in Waltz Mills, Pennsylvania									
CR-3 Crystal River Unit 3, Crystal River, Florida									
PRTR Plutonium Recycle Test Reactor, Hanford, Washington									
HTRE Heat Transfer Reactor Experiment									
ORR Oak Ridge Research Reactor									
MTR Materials Testing Reactor									
ETR Engineering Testing Reactor									
NRU Natural Uranium, Heavy Water Moderated and Cooled Test Reactor, Chalk River, Ontario									
SPERT-1									
U-A1	Wet	No	<0.01%	<0.79%	7%	a	Very small		31
UO2	Wet	No	<0.01%	a	0.06%	a	0		165
UO2	Wet	No	<0.01%	a	0.06%	a	0		155
BORAX-1	Wet	No	a	a	a	<1	<9.2		135

SNAPTRAN Systems for Nuclear Auxiliary Power Transient
SPERT Special Power Excursion Reactor Test

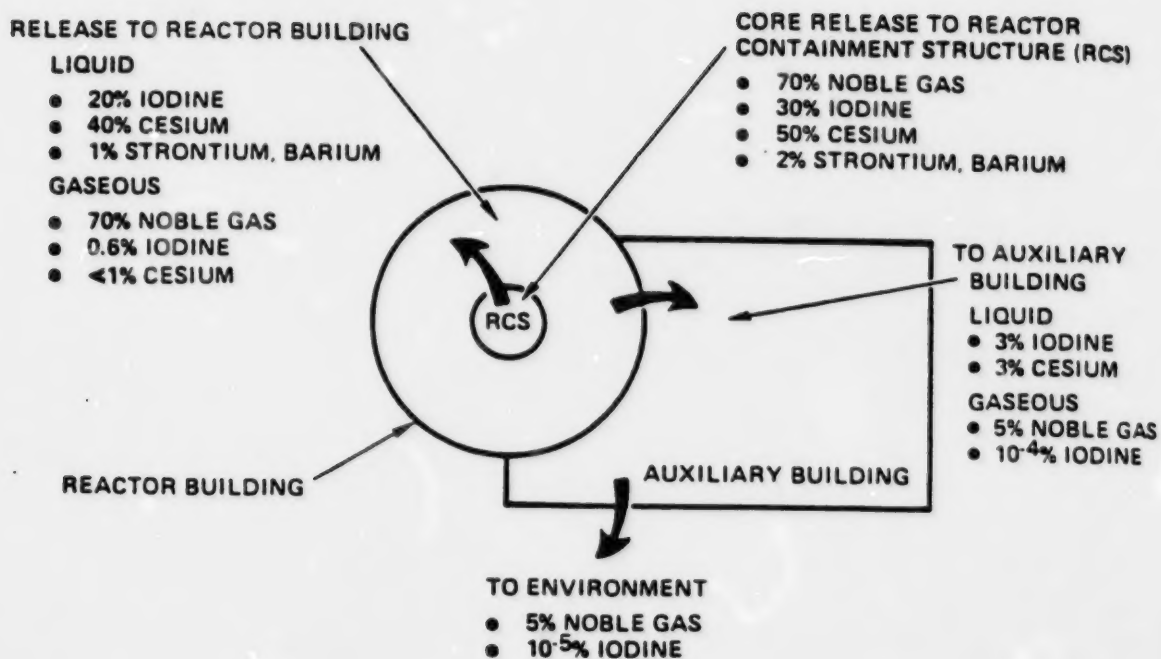


Figure A-1 Release fractions at TMI-2

from Morewitz, Fission product and aerosol behaviour following degraded core accidents, Nuclear Technology, 53, pp120-134, May, 1981

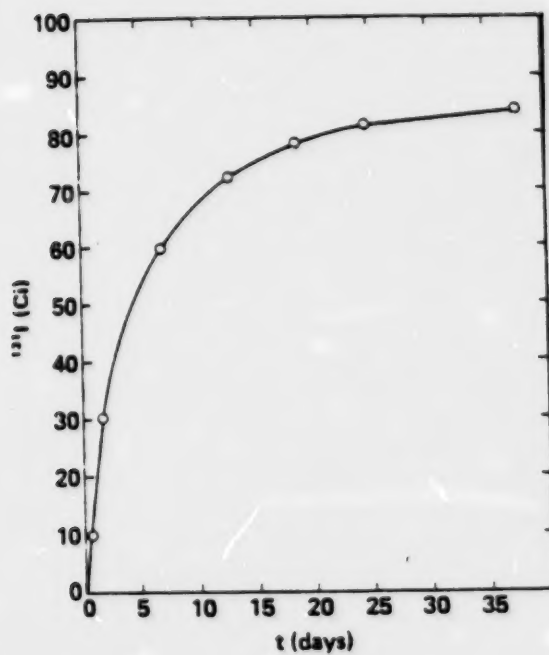


Figure A-2 Cumulative iodine release to the environment from the SL-1 accident.

from N.F. Islitzer, The role of meteorology following the nuclear accident in southease Idaho, IDO-19310, US Atomic Energy Commission, 1962.

was tracked out of the reactor building during the rescue operation. In total only about 0.01% of the fission products escaped from the leaky building although 5 to 10% had escaped from the reactor.

SNAPTRAN DESTRUCTIVE TESTS

Systems for Nuclear Power (SNAP) was a uranium-zirconium hydride, NaK cooled reactor designed for space applications. The SNAPTRAN series of destructive tests was conducted with unirradiated cores in the open atmosphere to evaluate the risk of launching the reactor over water and land. In the SNAPTRAN-2 test no water was present and 70% of the iodine, 75% of the noble gases and 45% of the tellurium were released to the atmosphere following a rapid reactivity insertion. In contrast, when a similar destruct test was conducted with the reactor submerged in an open tank of water, only noble gases were released to the atmosphere, but the other fission products were all retained in the water, in the fuel or plated on the pipes.

CRYSTAL RIVER-3 INCIDENT

This PWR underwent an incident when steam and 40,000 gal of contaminated primary coolant water were discharged on to the floor of the closed air-filled containment building. The water contained both I 131 and Xe 133. The Xe 133 was immediately released to the containment atmosphere but the I 131 was retained in the water to a large extent. A small amount of I131 was released to the containment atmosphere so that the amount peaked in about three days (fig A-3).

Windscale 1 ACCIDENT

The Windscale 1 reactor was a once-through air-cooled uranium-graphite plutonium production reactor. When the uranium metal fuel overheated and the graphite caught fire, fission products were released to the air stream discharging through a 400' stack over a two day period. The released iodine represented 12% of the available iodine inventory. It was originally suggested that the stack filters removed some 20 to 50,000 curies of iodine but further studies indicate (Chamberlain, 1987) that the filters removed very little of the iodine. The latest estimates suggest that 27,000Ci of I 131, with about 200 Ci of Sr 90, 2000 Ci of caesium 137 and "several hundred curies of Po 210" (Chamberlain, 1981) were released. From a public health point of view this was the worst reactor accident until Chernobyl. The radioactive plume was detected as far away as Norway and Egypt.

KIWI-TNT DESTRUCTIVE TEST

This test was conducted in the open atmosphere and used an unirradiated uranium-graphite rocket reactor (KIWI-B4) with air filled coolant passages. The reactor was deliberately destroyed by introducing a \$2000/sec reactivity ramp which vapourised 5 to

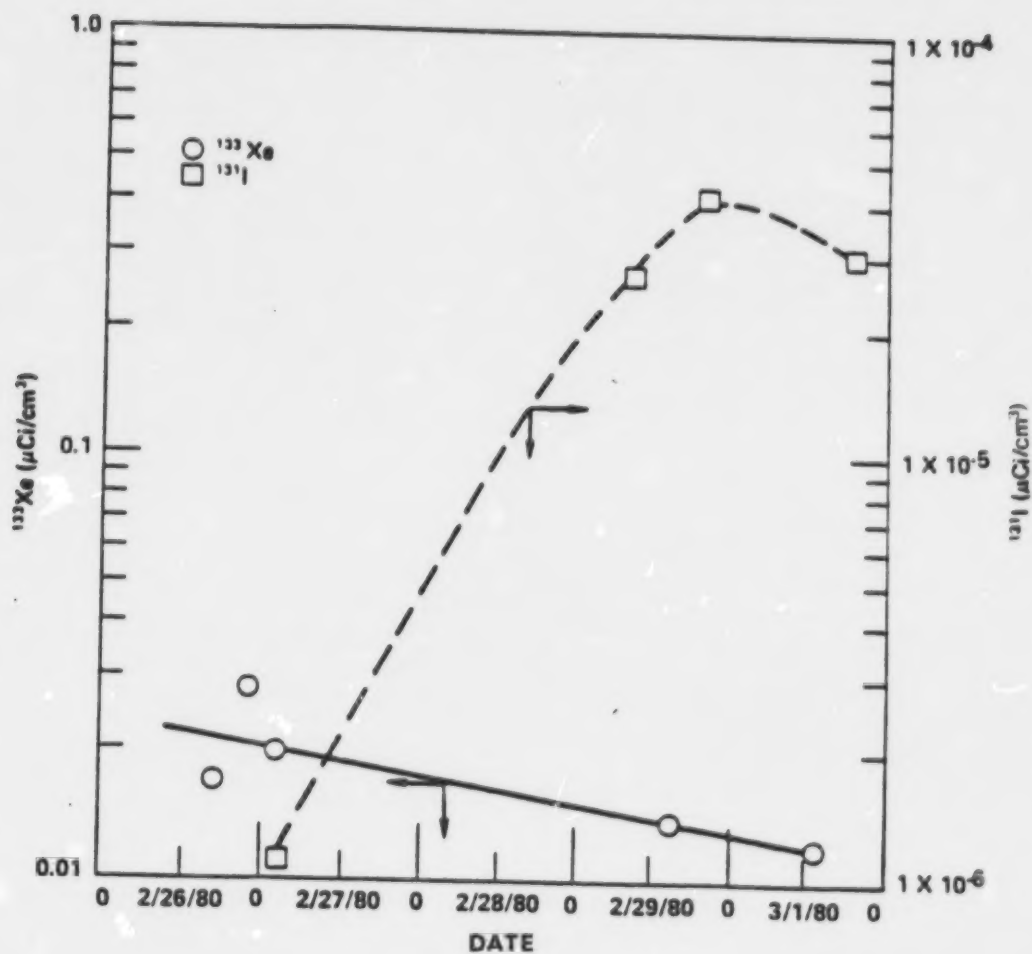


Figure A-3 Concentration of iodine 131 and xenon 133 in the containment atmosphere following the Crystal River-3 incident

from Analysis and evaluation of Crystal River Unit 3 Incident, NSAC-3/INPO-1, Nuclear Safety Analysis Center, Electric Power Research Institute, 1980

20% of the core and released 67% of the fission products to the effluent cloud. The blast was equivalent to the detonation of 150-200 lb of TNT. 67% by weight of the support material in the core was collected, while only 12% of the loaded fuel element material was recovered. The material in the cloud was mostly unfractionated fission products with the iodines still largely in the gaseous state. Tellurium and barium-lanthanum were detected as fine solid particles in the cloud. The radioactive plume was followed for more than 250 miles.

HTRE-3 ACCIDENT

The HTRE-3 facility was a once-through, air-cooled, zirconium hydride moderated reactor designed for aircraft propulsion. It used fuel elements of uranium dioxide powder dispersed in 80Ni-20Cr alloy. The reactor underwent a power excursion that partially melted about 68% of the core and released about 400 Ci (0.1%) of fission products from the facility stack to the atmosphere, depositing 34Ci of iodine isotopes and 0.1Ci of Sr 91 (sic) in a narrow sector downwind for 5.5 miles.

SPERT-1 DESTRUCTIVE TESTS

The SPERT-1 facility was designed so that destructive tests could be performed on a variety of water moderated reactor cores. The test took place in an open water tank housed in a rectangular building with a portion of the siding and roof removed prior to each test.

In the first test series an aluminum alloy, plate type, highly enriched core was used. Reactivity additions were successively increased, culminating in a 35Mwsec excursion that partially melted 35% of the core and dispersed the melted fuel. Approximately 7% of the noble gas inventory was released to the atmosphere. No iodine was detected in the atmosphere and it was all retained in the water.

Two additional destructive tests (155 and 165 Mwsecs) were performed with a core of stainless steel clad uranium dioxide. In each test two fuel rods ruptured and dispersed 2kg of 600 C, 100 micrometre sized uranium dioxide particles into the water. Only noble gases escaped to the atmosphere, while the iodine was retained in the water, as were the other fission products.

PREDEFECTED FUEL EXPERIMENTS

Experiments in pool type reactors showed that only 0.01% of the iodine released from the fuel penetrated to the atmosphere above the pool. In another case of fuel melting, 1000 Ci of fission products were released to the coolant but only 150 to 200 mCi of iodine was released to the atmosphere through the stack scrubber and filter. The peak iodine release was on the second day.

CHEMICAL COMPOSITION OF FISSION PRODUCTS

Adamson (1976) in a study of the behaviour of volatile reactive fission products in LMFBRs concluded from thermodynamic data and out of pile experiments that iodine will migrate exclusively as CsI(v) . Deposition of CsI(s) will occur at the cladding inner surface, but "even at temperatures of 550 to 750 C its vapour pressure is sufficiently high that it will tend to migrate axially toward blanket and plenum zones". Tellurium is also known to migrate down temperature gradients accompanied by caesium in oxide environments. Adamson reports that the major portion of fission product caesium released from hot fuel is transported down the radial temperature gradient as Cs(v) or, under certain conditions, as CsOH .

Malinauskas (1980) reported results of out of pile tests in which previously irradiated LWR fuel pins were heated in a horizontal furnace with an attached thermal gradient tube. When the fuel cladding of the pin was purged with purified helium, iodine was released as CsI . In a steam environment CsI , CsOH and antimony were released with trace amounts of ruthenium. In dry air both caesium oxide and molecular iodine were released with significant amounts of ruthenium, probably as ruthenium tetroxide.

According to Morewitz there is a chemical explanation for both the limited release of iodine and the delay in releasing it when water is present. First, iodine is present in the intact fuel as caesium iodide and not as molecular iodine, because there is ten times as much caesium produced in fission as iodine and, since both are volatile, they migrate to the cooler fuel regions where they combine. The chemical form of iodine in the fuel (caesium iodide) has been confirmed by tests (Adamson, and Cubicciotti and Sanecki). Caesium iodide is one of the most stable compounds of iodine but it easily dissolves in water to form caesium and iodine ions. The only way to obtain gaseous forms of iodine (iodine vapour, hypoiodous acid and methyl iodide) from a water solution of caesium iodide is either to oxidise the ions slowly by contacting the water surface with air, or to form organic iodides by slowly reacting the ions with hydrocarbon materials (paint, etc). If the solution is made basic (by the addition of NaOH or CsOH for example) the oxidation of the iodine ion is largely prevented. Most of the other volatile fission products are also very soluble in water. Rubidium, strontium, barium, caesium and their oxides dissolve in water after first reacting to form the respective hydroxides. Arsenic and selenium oxides are directly soluble in water.

CONCLUSION

Environmental releases of fission products from LWR accidents have been very small fractions of the available inventory and, in the case of iodine occurred only slowly. In no instance did the release of iodine exceed 0.5% or occur over a period of less than three days. Since the radioactive releases from LWR accidents will be small and slow, it is clear that severe measures such as

rapid evacuation of the nearby population or the issue of thyroid blocking agents are not required.

However in reactor accidents where water was absent and the containment poor, the release of fission products to the environment was large and rapid. the differences in release fractions and rate are related to the physical and chemical states attained by the fission products under the accident conditions.

A2.2

CHARACTERISATION OF DEPOSITS ON INSIDE SURFACES OF LWR CLADDING

by Cubicciotti and Sanecki J Nuc Mat 78 (1978) 96-111

This paper describes examination of Zircaloy cladding from uranium dioxide fuel rods irradiated under two different regimes (one resulting in significant fission gas release and solid fission product deposition on the walls, and the other involving high burn up at low power with almost no fission gas release), to determine the nature of the deposits on the inner wall of the cladding. The likely method of transport of fission products to the cladding was by the vapour.

Two of the rods examined were from pellet-cladding interaction (PCI) failures in unpressurised fuel rods from Maine Yankee Core 1, irradiated to 13GWd/Mg(U) at linear power less than 300W/cm. The rods showed high (10-13%) fission gas releases and peaks of activity from Cs137 at positions corresponding to pellet interfaces. The deposits were of three types: (i) sharply defined linear stripes which occurred opposite cracks in the fuel (ii) concentrations of more massive deposits at the locations of pellet-pellet interfaces and (iii) circular mound like deposits at random locations. In addition there were occasional chips of fuel that strongly adhered to the cladding. The pieces of cladding were examined in a scanning electron microscope equipped with an energy dispersive X-ray analyser. The high radiation field broadened the X-ray fluorescence energy peaks (in the case of unirradiated vs irradiated Zircaloy the half width of the peak was broadened from 170 to 500ev). A typical linear deposit consisted of two parallel gray bands about 0.1mm apart. There seemed to be a concentration of particles of random size and shape between the bands. The gray bands consisted of nodules around 1 to 2 micrometres in diameter, separated by about 1 micrometre, firmly attached to the substrate which was presumably a surface coating of zirconium dioxide on the Zircaloy. Analysis of these nodules showed mainly Zr (presumably due to the penetration of the electron beam through the nodules) and some U, Cs and Te. Some almost white angular particles (5 to 10 times larger than the nodules) contained U and Cs and were probably a caesium uranate.

Particles observed between the bands included pieces of uranium dioxide, particles of caesium uranate that were sometimes crystalline and sometimes amorphous, and some amorphous deposits whose analysis showed Cs and Te only and which were presumed to be caesium telluride. Occasionally small amounts of Fe, Mn, Cu, and Cl were observed in some of the amorphous deposits (possibly from sample contamination or from specimen mounting stubs). Some cubic crystals were observed in a linear deposit and showed an X-ray spectrum very similar to that of a chemically pure crystal of caesium iodide if allowance is made for spectral broadening due to the high radiation field. (it should be noted that crystals of caesium iodide are rhombic rather than cubic). Material observed in the central region of another linear deposit showed particles

of uranium dioxide with various amounts of zirconium dioxide and some features showing various ratios of U,Cs,Te,Zr with some Mn,Cu and Cl.

H.R. ROBINSON ROD

The H.R. Robinson rod had been irradiated to an average burnup of 28 Gwd/Mg(U) at an average power of 230 w/cm. Axial gamma scans showed no Cs peaks and fission gas release was small (<1%). The inner surface of the cladding was relatively clean and free of deposits. None of the features of the Maine Yankee cladding (stripes opposite fuel cracks, pellet interface deposits or circular mounds) was observed. No fission products were detected except for an occasional trace of caesium.

The chemical states of the fission products in uranium dioxide fuel are not well known. A first assessment as to whether an element would be expected to be oxidised was made by Cubicciotti and Sanecki on the basis of data on free energies of formation of pure solid metal oxides, although when the fission products are incorporated in uranium dioxide they may have different oxidation states and solid solution states which occur at different oxygen potentials. It is expected that because of the great stability of the halides of caesium, bromine and iodine will react with caesium in preference to oxygen. Compounds such as CsI, RbI, CsBr, RbBr and caesium telluride are sufficiently volatile that they might be expected to be vapour transported from the fuel during normal operating temperatures (1500K at the centre to 1000K at the outer surface). However the release of even the highly volatile noble gases does not occur during normal rod operation even after relatively large burnup. Cubicciotti and Sanecki believe that the formation of deposits such as CsI and caesium telluride opposite fuel cracks result from the release of the fission products from the fuel centre when its temperature exceeded a certain threshold value, reaction of released atoms in the gas phase to form stable molecules, transport of these along fuel cracks and deposition on the cladding which is the coolest surface available. Only small amounts of fission products were deposited on the H.R. Robinson cladding. Since only very little of the rare gases had escaped, the Cubicciotti and Sanecki model suggests that this was because they had not been released from the fuel, not because their volatilities were too small.

Note: Cubicciotti and Sanecki are often quoted as the authority for the statement that the chemical form of most of the iodine released from an accident to a water reactor will be as caesium iodide. e.g. Bruce B Safety Analysis section 3.5.2.5.4.2 "IODINE BEHAVIOUR IN FUEL Iodine produced in the fuel reacts chemically with metallic fission products such as tellurium and caesium forming metal iodides. Indeed caesium iodide (CsI) has been identified as the DOMINANT SPECIES OF IODINE INSIDE IRRADIATED FUEL" and a reference to Cubicciotti and Sanecki is given. In fact, C & S say nothing of the sort. They are looking at deposits on the inside of the cladding, NOT inside the fuel. only a small part of their paper even refers to caesium iodide

and they NEVER even imply, much less say, that CsI is the dominant species of iodine. The authors of the Bruce B Safety Analysis then go on immediately to say "DUE TO THE STABILITY AND LOW VOLATILITY OF METAL IODIDES AT HIGH TEMPERATURES, A LARGE RETENTION OF IODINE IN THE FUEL IS EXPECTED" This confirms that they believe that CsI exists in the fuel matrix, which is never even suggested by Cubicciotti and Sanecki and, what's more, that they are relying on this 'fact' to some extent in their safety analysis.

Further in the Pickering Restart Analysis (3.1.2.6,p84) "It is now widely recognised that IODINE IS RELEASED FROM THE FUEL AS READILY SOLUBLE AND CONDENSIBLE CsI....."

A2.3 IN VESSEL RELEASE OF RADIONUCLIDES AND GENERATION OF AEROSOLS

by R.R. Hobbins, D.J. Osetek and D.L. Hagrman, IAEA SM 281/7, 1986

Abstract: Review of state of knowledge (experimental and theoretical) of release of radionuclides and generation of aerosols from reactor pressure vessel under severe accident conditions. Recent experimental results on release of noble gases, iodine and caesium compared with NUREG 0772 values which tend to form an upper bound to experimental data. The considerable scatter in data on volatile fission product release can be understood in terms of several phenomena e.g. fuel morphology and fission product distribution as a function of burnup and fuel oxidation. Holdup, of tellurium in Zircaloy cladding and the subsequent release of tellurium on Zircaloy oxidation are qualitatively understood. Determination of the chemical species of fission products released is an important area which is currently being studied. Much work remains to be done to understand fission product/aerosol chemical interaction.

In trace-irradiated fuel the noble gases, iodine and caesium probably diffuse as individual atoms and in higher burnup fuel they most likely are swept out in bubbles containing a mix of species.

The scatter among data on the release of volatile fission products is quite large, covering about three orders of magnitude, with the NUREG 0772 curves forming an upper bound. Highest release rates tend to be associated with highest burnups: the effect of fuel oxidation is also quite strong. There is also an enhanced release on fuel cool down which is attributed to boundary microcracking.

Preliminary results on the fission product content of the TMI core indicate release fractions of 44% for ruthenium, 80% for iodine and caesium, 77% for antimony, 6% for strontium and essentially zero for cerium. Microstructures revealed that temperatures up to fuel melting (3100K) occurred in the core, but the debris also contained much fuel that had experienced much lower temperatures (1900K). Relatively large release of ruthenium from the TMI core debris is unexpected.

There is little experimental information available on the chemical form of fission products released from fuel. Work is underway at Argonne to measure chemical species released from irradiated fuel specimens as a function of temperature using a mass spectrometer. Similar techniques employed at Batelle Columbus on the effect of high pressures on release. EARLY RESULTS FROM THE ARGONNE EXPERIMENTS HAVE IDENTIFIED ONLY THE ATOMIC SPECIES OF IODINE, CAESIUM AND TELLURIUM AT TEMPERATURES UP TO 1773K IN FUEL IRRADIATED TO A BURNUP OF 31gwD/t. Raman and luminescence spectroscopy have been used at the Idaho National Engineering Laboratory to investigate the chemical forms deposited on surfaces in the effluent handling system downstream of the fuel in the PBF-SFD tests. TO DATE CsI HAS NOT BEEN IDENTIFIED.

Fission products are incorporated into the fuel either in elemental form or as oxides, although other compounds such as CsI and uranates are possible. Chemical compounds formed within the fuel may be soluble in the uranium dioxide lattice. Those that are soluble allow the possibility of ion diffusion within the lattice BUT soluble species have much reduced volatilities. Insoluble molecules such as BaO are essentially immobile because of their size, although mobility via vapourisation into bubbles is a possibility. Some fission products (e.g. ruthenium) tend to form insoluble metallic ingots in moderately high burnup fuels and have transport limitations similar to large molecules. RADIONUCLIDES EXPECTED TO BE IN ATOMIC FORM WITHIN THE FUEL ARE NOBLE GASES, IODINE, CAESIUM, TELLURIUM, ANTIMONY AND RUTHENIUM. Soluble oxides are formed by lanthanum, cerium and zirconium. Insoluble oxides are formed by strontium and barium. Much work remains to be done on the determination of chemical species released from the fuel and the modelling of the migration of the species within the fuel.

An area that remains relatively poorly explored but that is potentially important is chemical interaction between fission product vapours and aerosols. This could be significant because of the high temperatures and large surface areas of the aerosols and could affect the source term by changing the deposition/revaporisation behaviour of certain radionuclides during transport in vessel and in primary system piping.

It is generally recognised that aerosols generated in-vessel can interact with fission product vapours and play a significant role in the subsequent transport and, possibly, deposition of fission products. Aerosols can be generated from control materials, structural materials, fuel rod materials and fission products. THE GENERATION OF AEROSOLS IN-VESSEL IS ONE OF THE LEAST UNDERSTOOD AREAS IN SOURCE TERM SCIENCE. Wichner and Spence (1985) assumed that the equilibrium vapour over a molten core is condensed into aerosol material. The result is a quantity of aerosol equivalent to about 1% of the core mass. It is expected that transient effects such as downward flow and freezing of liquid materials will reduce the aerosol generation well below this estimate.

Out of pile electrically heated experiments at ORNL have indicated that the largest contributor to aerosol production up to 2000K is cadmium from cadmium-silver-indium control rods. In-pile tests without control rod material at Sandia have shown tin to be the major contributor to aerosol.

A2.4 STEP (SOURCE TERM EXPERIMENTS PROJECT)

(information abstracted from EPRI NP-4967M, Interim Report, Nov 1986)

STEP was started in 1982 by a consortium (EPRI, Ontario Hydro, DOE, NRC, Belgonucleaire) to obtain data on the characteristics of fission products and on the components of structural materials which might be released from reactor fuel under degraded core accident conditions. It was designed to obtain data on the physicochemical properties near the point of origin, of biologically important volatile fission products released by fuel rods early in severe LWR accidents. Samples of aerosol were collected on coupons of different materials hung from a sample tree in the flow of steam.

After the tests the coupons were examined by scanning electron microscopy and energy dispersive X-ray analysis.

The results showed that in low pressure high fuel heat up conditions the deposition samples consisted primarily of tin from the cladding together with caesium, molybdenum and rubidium. Iodine (often in this study associated with caesium) and tellurium appeared less frequently.

In tests at high pressure and low fuel heat up rates the deposits contained mostly iron and silicon from the experimental apparatus together with some molybdenum and caesium.

Fuel and cladding fragments appeared in the high heat up rate samples, but only cladding fragments at low heat up rates. Iodine and tellurium were not observed in samples from tests at low heat up rates.

Particulate material in the deposits ranged from less than micrometre (diameter?) particles to agglomerates as large as several micrometres. Deposits containing molybdenum, caesium and iodine were generally amorphous, although some were definitely crystalline in nature.

Particularly noteworthy is the identification of molybdenum as an important component since this is not usually regarded as a volatile fission product (MP 2617 C, BP 4612 C)

Altogether four in-pile experiments were conducted at the Argonne National Laboratory transient reactor test facility in Idaho.

It was noted at Three Mile Island that the quantities of caesium and iodine released to the environment were very much lower than predicted from conventional reactor accident analysis. It was postulated that this was due to the presence of the water coolant. Iodine is believed to combine with caesium in the fuel forming CsI. This is highly soluble in water (44g/100ml in cold water, 160 in hot). It is postulated that condensing steam would dissolve the CsI, also that CsI has a high dew point and would

therefore condense to form aerosol particles in the cooler regions of the reactor, where they would be captured by deposition on surfaces during transport (NB depends greatly on particle size distribution and flow conditions). It is stated that the deposition processes are enhanced in humid conditions (diffusiophoresis?). The predominant form of caesium is expected to be caesium hydroxide, CsOH , which is highly soluble (395g/100ml in cold water) and also has a high dew point. (MP 272 C)

The purpose of the STEP experiments is to obtain data on release rates and fission product behaviour in the presence of steam and it is hoped that these, in conjunction with the development of computer simulation codes will yield sufficient data for the formulation of realistic source term standards. It is necessary to define (a) the magnitude and release rates of fission products from degraded fuel pins, (b) the physical and chemical characteristics of the released fission products (c) transport mechanisms of the volatile and condensed fission products from the core to the reactor internals (?) and (d) transport mechanisms through postulated breaches in the primary and secondary containments to the environment. The STEP experiments should provide data on (a) and (b) and provide information on aerosol formation and transport which can be applied to (c) and (d). The experiments were conducted 'in pile' so that the heating of the fuel element was nuclear and steam was introduced into the experimental arrangement at the temperatures and pressures postulated for accident scenarios, so that fission products were released under conditions closely resembling those expected during an accident.

The accident sequences selected for the four STEP tests covered a range of accident scenarios based on risk dominance, combining factors relating to environmental hazard and probability of occurrence. The three primary experimental parameters were pressure, fuel heat up rate and the presence or absence of control rod material. The pressure of the steam in the reactor core at the time of release influences the chemical and physical behaviour of the fission products and hence their transport characteristics. Fuel heat up rate determines the migration of the fission products through the fuel matrix and hence affects the timing and composition of the release. Volatile materials introduced by disruption of control rods also affects the chemical and physical behaviour of the fission products.

The experimental set up was designed to subject pre-irradiated fuel to the temperatures, heat up rates and steam flows predicted by computer analysis of selected accident scenarios. The object of each test was to release sufficient fission products for characterisation by ensuring adequate fuel at temperature times, thus causing extensive cladding disruption by the combined effects of oxidation by the steam, hoop stress failure and melting. Temperatures in the range 850 C-950 C are required for cladding rupture by internal pressure. Cladding melting is expected to occur at about 1825 C. Thus maintaining cladding temperature above

1825 C was felt to be sufficient to ensure fission product release from the fuel-cladding gap as well as from the fuel matrix. The high fuel temperatures were induced by nuclear heating and from the heat of reaction of the cladding oxidation. Fuel melting (about 2800 C) was avoided. Identical systems were used in each of the tests.

Electrical heaters were used to ensure that no liquid water was present in the system (temperature maintained at 370 C). Fuel rods used were from BR3 (Belgonucleaire), 1m long. Zircaloy 4 cladding had OD 9.5mm with a 0.65 mm wall. Rods were typical PWR rods except a factor 3 shorter and higher 235 enrichment and had been irradiated in BR3 to a burn up of 3 to 4.5 atom per cent. Origen (2) calculations of fission product content of rods are:

FUEL ROD FISSION PRODUCT INVENTORIES

Element	APPROXIMATE MASS (G/ROD)		
	STEP 1	STEP 2	STEP 3/4
Cs	1.29	1.15	1.30
Mo	1.65	1.54	1.79
Rb	0.21	0.19	0.22
Sn	0.015	0.014	0.017
I	0.082	0.076	0.090
Te	0.18	0.17	0.20
Zr	1.96	1.84	2.13
Ag	0.010	0.009	0.012
Cd	0.013	0.012	0.016
Ru	0.87	0.80	0.95
Sb	0.004	0.003	0.004
Ba	0.80	0.75	0.89
Sr	0.50	0.46	0.53
Ni, In, Cu	Negligible		

No significant amounts of long lived fission products were created during the TREAT irradiations.

Aerosol characterisation system

A fraction of the particulate and gaseous fission products entrained in the steam flow was collected on devices attached to a type 304 stainless steel tube hanging above the fuel elements. This sample tree contained three sample stations approximately 1.1, 1.9 and 2.7 metres above fuel mid height.

Each station contained a tab assembly, a condensate trap and two thermocouples. The tab assemblies had eight tabs suspended into the flow. Two 0.64 cm square sample coupons were tied to the tabs (one on each face) with platinum wire. Four of the tabs

were perpendicular to the flow and four parallel. It was intended that post-test examination of the particulates and condensed and sorbed vapour species on the coupons would provide information on the chemistry of the fission product laden steam.

The cone shaped (304) stainless steel condensate traps were designed to collect liquid which condensed and flowed down the sample tree tube.

Aerosol sampling canisters

Approximately 13% of the fission product laden steam/hydrogen mixture was directed into two aerosol sampling canisters. The canister inlets were situated 1 and 2.6 m above the fuel mid height. Each canister consisted of three chambers with 14 collection stages per chamber. The stages were partitioned to create a labyrinthine channel giving a flow path 3.18m long in a space which was only 0.16m high and 5.9cm in diameter.

The stages contained settling plates which collected particles by gravitational settling and diffusion and fine wires which collected particles by impaction, interception and diffusion. Gaseous species, if present, could condense or be sorbed on to these devices. The settling plates were on the floor of the stages and the fine wires suspended perpendicular to the flow. Coupons of various materials were located on the floors of the stages and the wires were made of a number of materials. It was intended to analyse these collection devices primarily by scanning electron microscopy and by energy dispersive X-ray analysis. The collected particles would be counted to determine particle size and mass distributions.

Fission product detection system (FADS)

Filters located in the lines downstream of the canister chambers collect a portion of the fission products not removed in the canister stages. The filters consist of three layers of stainless steel mesh with decreasing pore size. They had a rating of 2 micrometre nominal (i.e. they would collect 95 to 98% of 2 micrometre particles incident on them). The projected face area of each filter was 6.4 sq cm. The system was intended to measure the accumulation of radioactive fission products using gamma ray spectroscopy to obtain real time identification of the radioisotopes making up the aerosol particles.

Main effluent filter

The main effluent filter was designed to capture the majority of particles escaping from the primary vessel and was made of pleated woven type 316 stainless steel. It was again rated at 2 micrometre nominal and had a surface area of 650 sq cm. The filters in STEP 3 and 4 had a prefilter consisting of a 1.2 cm thick annular bed of 3mm diameter silica beads surrounding the pleated metal filter in the same housing.

TEST OPERATIONS

The test parameters which were varied were pressure, fuel heat up rate and the presence or absence of control rod material. Complete coverage would have required eight tests, but only four were performed. The possible accident sequences selected for study were AD, TQUW and TMLB (from WASH-1400).

STEP 1 (AD)

Corresponds to a large coolant pipe break in a PWR with failure of emergency core cooling, rapid depressurisation and prompt core uncovering. In this test the energy deposition rate was 3.4 MJ/Kg, the test lasted for 20.6 mins, the pressure was 0.32 MPa and the initial heat up rate 1.7K/sec.

Fuel disruption was suggested by the observed temperatures and verified by post test neutron radiography and by the large amounts of fission products in various system components. The radiation levels required that examination of the samples be postponed for a month after the test and during this time the aerosol characterisation system was exposed to water vapour at ambient pressures (vp 0.0025 MPa or 0.36 psi). There was no condensation in the primary.

The coupons had deposits of sub micron to micron size particles and sub micron to super micron 'non particulate masses' with loadings heavy enough to obscure the substrate almost completely. Irregular particles composed of tin and caesium with lesser amounts of molybdenum and rubidium were most common. These particles did not appear to have a crystalline structure. Smoother geometric particles and masses (implying crystalline structure according to the report - could it not imply melting?) were observed with higher proportions of caesium and molybdenum - the irregular particles had higher concentrations of tin. Deposits looking like 'splatters' also showed higher Cs and Mb concentrations. Caesium rich needle shaped particles were also observed.

Iodine was not detected on the upper or lower coupons. During an initial examination iodine with caesium was observed on the middle coupon in the elongated structures on splatters but could not be located again when the energy spectra were being recorded.

Super micron fluted rods of zirconium based material were seen on all three coupons (to a lesser extent on the upper coupon). Smooth spherical particles, similar in size, were seen on all coupons; their constituents (identified only on the upper coupon) were predominantly zirconium. Their shape implies formation by condensation and transport as aerosol particles. Super micron sized uranium deposits identified on lower and middle coupons and, possibly, on upper coupon, apparently fuel fragments.

Zirconium based rods, uranium fragments and spheres have micron and submicron particles on them. Tellurium observed on upper coupon, but identification questionable. Antimony possibly detected on upper coupon.

STEP 2 (TQUW)

A transient event in BWR combined with failures of high pressure emergency core cooling and long term decay heat removal. Automatic depressurisation and temporary low pressure coolant injection results in a very long time to core uncovering. The energy deposition was 2.9 MJ/Kg and the test lasted for 20.5 min. The planned pressure for the test was 0.14 MPa. It started at 0.16 MPa and a pressure rise began 8 min into the test. At 0.97 MPa the reactor was manually shut down and pressure continued to rise to 1.24 MPa when steam supply was shut off. The initial heat up rate associated with the TQUW sequence was 0.7 K/s. The lowest rate attainable with the reactor and test vehicle constraints was 1.2 K/s. In fact the experiment rate was 2.4 K/s. The reason for this was apparently that oxidation of the cladding occurred considerably before melting. The experiment was terminated early because the pressure rose to the limiting value. The data suggested that the fuel disrupted and this was confirmed by radiography and by the detection of fission products outside the fuel region, although in lesser amounts than in STEP 1. Post test nitrogen purging and primary vessel isolation from the receiver tank occurred within a week after the experiment.

The STEP 2 coupons had relatively uniform deposits, which were thin enough for the substrate to be visible. Most of the deposits consisted of irregular particles composed of tin and caesium with rubidium and/or silicon and molybdenum. Larger particles appear to be agglomerates. Larger irregular masses of agglomerated material were also observed on each coupon. High magnification photographs showed that the smallest distinct unit of visible material was an irregular particle. It is possible that particles smaller than 0.25 microns were present but these are difficult to distinguish from the substrate. Smooth geometric particles (like STEP 1) but more cubic were found on the lower coupon and the predominant component of them was caesium. Smooth rods found on each coupon were similar in appearance but those on the lower two coupons contained caesium, iodine and silver while on the upper coupon they were silicon and caesium based. The middle coupon had more of these long deposits than the other two. The caesium, iodine and silver deposits on the upper coupon are long and thin, but have a flatter appearance than the rods on the lower two coupons. Very regularly shaped flat rectangular deposits containing caesium and iodine were also seen on the upper coupon. The ratio of the Cs/I peaks is consistent with that for CsI for all the deposits except for the long caesium, iodine and silver deposits on the upper coupon where the iodine peak is higher than for CsI. A clustered deposit made up of smaller rods was found on the middle coupon containing caesium, uranium, rubidium and molybdenum - quite different from the other elongated smooth deposits.

Fluted rods were found on all three coupons, fewer on the middle coupon. Those on the upper coupon contained zirconium and were consistent with those from STEP 1. Smooth spheres were found on all three coupons with zirconium as the dominant constituent - consistent with STEP 1. Uranium fragments were found on the upper and lower coupons (latter not definite). Small particles deposited on rods and fragments though not as many as in STEP 1. An unidentified thin layer (appears to be frozen liquid) on lower two coupons. Unidentified long fibre like deposits on upper coupon. Tellurium and antimony NOT observed.

STEP 3 (TMLB)

This PWR fault was a transient with failure of main feedwater and auxiliary feedwater and failure to recover electric power; no depressurisation, coolant loss by venting through relief valves, core uncovering in one hour or more. The rate of energy deposition was 3.2 MJ/Kg, the duration of the test 21.4 mins, the pressure 8 MPa and the initial heat up rate 0.6 K/s. Fuel disruption and fission product release were confirmed. Post test nitrogen purging and primary vessel isolation from the receiver tank occurred within a few days of the test.

The deposits on all three coupons consisted of a relatively uniform layer of spherically shaped material of diameters varying between sub micron and several microns. High magnification images of material on the middle and upper coupons showed that the spheres were composed of clusters of slivers. In addition there was a relatively uniform layer of needle shaped material on the lower coupon. The middle and upper coupons have layers of individual slivers which appear to be similar to those making up the spherical material. The clusters of slivers and needle shaped deposits suggest crystalline structures and the possibility of growth on the surface of the coupons rather than formation in the gas stream and subsequent deposition.

The spherical material consisted primarily of iron with lesser amounts of rubidium, caesium, silicon and tin present. The ratio of iron to nickel and chromium peaks in the X-ray spectra is significantly higher than that for stainless steel indicating that the iron signal is from the deposit.

Irregularly shaped deposits seen at low magnification on all three coupons with more on the lower coupon. Higher magnification revealed agglomerated structures containing silicon, caesium, iron, tin, molybdenum, copper. Other smaller irregular deposits on the lower and middle coupons not identified but shown by backscattered electrons to be composed of heavy elements. Fluted rods containing zirconium (similar to STEP 1 and 2) were seen only on the middle coupon which also contained at least one spherical particle composed partly of zirconium and resembled those seen in STEP 1 and 2. No iodine, tellurium, antimony or uranium was detected.

STEP 4 (TMLB) as STEP 3 except control rod simulant included

Energy deposition was 3.6 MJ/Kg, the test lasted for 23.8 mins. Pressure was 7.86 MPa and the initial heat up rate 0.5 K/s. STEP 4 contained a 2.5mm rod simulating a Ag/In/Cd control rod in mid life (i.e. with tin). The behaviour of the test was similar to STEP 3. Fuel and control rod disruption and fission product release were confirmed.

The deposits on the coupons were primarily a layer of smooth geometrically shaped particles, sub micron to micron in size, mainly iron and silicon on the lower coupon and cadmium, molybdenum and silicon on the upper coupon. Particles do not appear to be composites of crystalline material. Unidentified sub micron needles observed on middle and upper coupons; the heavy layer of particles on the lower coupon may conceal similar needles.

Also larger irregular particles on each coupon, identified as silicon on the lower and middle coupons. Iron definitely the main constituent of the small particles on the lower coupon.

Two sizes of Zr based fluted rods seen on the three coupons, roughly similar number density on each. Similar to fluted rods in other tests. Of the materials present in the simulated control rod material (silver, indium, cadmium and tin), only cadmium and tin were observed. Caesium not identified and tin not so prominent as in other tests. Iodine, tellurium, antimony and uranium not found.

The above information has been abstracted from EPRI NP 4967M dated November 1986. It gives no information regarding hydrogen generation or the results of examining samples other than the coupons and the implication is that the work is still in process. Surprisingly, a paper by Herceg et al presented at the IAEA Symposium 28 October to 1 November 1985 (IAEA-SM-281/11), a year earlier, gives more information on aerosol analysis and hydrogen generation, in particular specific size ranges for the particles captured on the wires in the aerosol samplers e.g. 'easily retrievable quick look deposition coupons were recovered from the sample trees of each vehicle and subjected to scanning electron microscopy. Elemental identifications were made via analysis of fluorescence x-rays generated during this process. A FEW OF THE AEROSOL CHARACTERIZATION SYSTEM DEPOSITION COUPONS AND FINE WIRE IMPACTORS FROM STEP 1 HAVE UNDERGONE SIMILAR EXAMINATION. (no mention of these results is made in EPRI NP-4967M.) 'Initial observations on these samples reveal the presence in the deposited material of the fission products Cs, I, Rb, Mo and Te; the cladding materials Zr and Sn, the probable structure constituents Fe, Mo and Si; the fuel U; and Ag, Cd and Cu from sources yet to be determined. Only the STEP 1 aerosol characterisation train samples have been subjected to detailed particle size counting and measurement, but tree samples from other tests appear to be

qualitatively similar. Two distinct particle populations appear to be present, one of sub micrometre size and the other generally larger than a micrometre and often several tens of micrometres in at least one dimension.

The smaller size particles outnumber the larger by many orders of magnitude. They appear to have been formed by nucleation from the vapour phase and contain primarily Cs, but also Sn, Rb, and Mo and occasionally Te. Cs and I are found together, often with no other constituents and sometimes with Ag (presumably from deposition sample coupons), in the larger particles, usually having an elongated and crystalline form. Iodine is found only in association with Cs on all of the samples examined to date. **ALTHOUGH MOLECULAR CsI HAS NOT YET BEEN UNEQUIVOCALLY IDENTIFIED, ALL AVAILABLE EVIDENCE POINTS IN THIS DIRECTION.**

Zirconium and U are usually found in larger particles and appear to have been released as fragments, possibly due to vigorous Zr oxidation. The variety of sizes and shapes of the particulate and non particulate deposits is thus far felt to be remarkable. In spite of this, many thousands of particles were counted by SEM imagery on several areas of STEP 1 coupons and impactor wires and the data processed to yield particle concentrations by size in the flowing steam-hydrogen mixture above the fuel. **A CONSISTENT PICTURE SEEMS TO BE EMERGING OF AN AEROSOL WITH MAXIMUM NUMBER CONCENTRATION IN THE 0.3 TO 0.5 MICROMETRE DIAMETER RANGE.**

A 2.5 RADIOIODINE CHEMISTRY IN REACTOR ACCIDENT CONDITIONS

by F. Garisto et al IAEA-SM-281/46, 1986

Detailed research has shown and TMI has confirmed that in water cooled power reactors potential radioiodine releases are far less than previously assumed and are regulated by the chemical behaviour of iodine.

Behaviour of iodine in the PHTS strongly influenced by presence of caesium. Cs is volatile at normal fuel temperature and accumulates in the grain boundaries and fuel cladding gap. $Cs/I = 10$ in the fuel. There is experimental evidence that CsI is formed in the fuel-cladding gap (Cubiciotti and Sanecki, 1978). THUS (!) caesium and iodine should be released simultaneously as CsI during the initial stages of a fuel failure. The reducing steam conditions in the PHTS during a LOCA (due to high PHTS pressure and hydrogen production by the zirconium-steam reaction) should prevent fuel oxidation. Fuel oxidation could lead to release of molecular iodine due to entrapment of caesium in the fuel by the formation of caesium uranates. The thermodynamic stability of CsI, compared to all other iodine species that can be formed in the PHTS ensures that for $Cs/I > 1$, CsI is the predominant iodine species under all the conditions (pressure, temperature, Cs/I ratio, hydrogen/water ratio) studied. On the other hand in a hydrogen/steam atmosphere, in the absence of caesium, HI is the predominant iodine species at all temperatures.

Both CsI and HI dissociate in contact with water to form the involatile iodine (-ve)ion. Therefore if CsI and HI are formed in the PHTS and contact water they will reach the containment vessel in an involatile form. Kinetic calculations show that for temperatures greater than 1000 K the dominant iodine species will reach a steady state in less than 0.1s. The worst case is an accident where all the caesium is released and reacts to form CsOH before the release of atomic iodine. Even under these conditions CsI becomes the dominant species in less than 0.01s.

The origin of the organic iodides formed at TMI is uncertain. According to Garisto et al the PHTS is not a source. They tend to the view that reaction with organic impurities is a likely cause. They believe that the most important airborne iodine during reactor accidents will be an organic iodide. Their conclusion is that insignificant quantities of airborne radioactive iodine would be released into the containment building.

A 2.6 LWR CONTAINMENT EXPERIMENTS (LACE)

The report LACE TR-012 (January 1987) describes the results of a series of experiments in which artificially generated aerosols (but which, in some degree, were thought to be representative of the aerosols which might be released in an LWR accident), were generated and released in quasi reactor containment conditions. The experiments were carried out at Hanford Engineering Development Laboratory. The tests were supported by a consortium whose members represented many international and national organisations including Commission of the European Communities, VTT (Finland), Ontario Hydro (Canada), CEA (France), BMFT (Germany), ENEA (Italy), JAERI (Japan), KEMA (Netherlands), UNESA (Spain), SKI (Sweden), EIR (Switzerland), AEA (UK), EPRI, DOE and NRC (USA). At an initial meeting held in Dec 1982 it was decided that the three accident situations which were most important in showing that a significant reduction in accident consequences could be obtained by demonstrating significant aerosol retention were (i) containment bypass sequences, (ii) containment leakage and failure to isolate and (iii) delayed containment failure (if aerosol resuspension were significant). It was considered that both soluble and insoluble aerosols should be used and that they should be made of materials that exist in LWR reactor cores. It was decided that the aerosols used should be formed by vapour condensation of materials in the core region of a reactor. MnO was chosen as the insoluble aerosol and CsOH for the soluble. An MnO aerosol was generated by vapourising metallic manganese in a plasma torch and allowing the vapour to react with steam to form MnO and the CsOH aerosol by vapourising elemental caesium in a furnace and allowing the vapour to react with steam to form CsOH. The two aerosol species were generated separately and mixed in a 5.2 cubic metre vessel to allow time for agglomeration and settling of oversize particles.

The planned series of tests was as follows

Test LA1 - Containment Bypass:

The objectives of test LA1 were to determine the retention and behaviour of aerosol materials flowing through a pipe to a vented downstream auxiliary building under severe accident conditions. the specific objectives were:

- measure the fraction of the aerosol contained in the pipe
- determine the mass fraction of aerosol as suspended particles and deposited on walls
- determine changes in aerosol characteristics resulting from flowing through the pipe
- determine aerosol behaviour in auxiliary building
measure aerosol mass fraction vented from auxiliary building

Test LA2 - Failure to isolate containment:

This was an integral test under conditions similar to those postulated for a severe LWR accident with containment isolation failure in which the magnitude of aerosol deposition within containment and leakage from containment are similar.

The specific objectives were:

- provide a data base of thermal-hydraulic measurements suitable for thermal-hydraulic computer code comparisons
- provide a data base of aerosol behaviour measurements in a containment with pre-existing leak paths, suitable for aerosol computer code comparisons
- determine the effect of leak path location (high and low) on aerosol mass leaked from the containment
- determine the particle size distribution of aerosol escaping the containment through orifice-type leak paths

Test LA3 - Containment Bypass:

The objective was to determine the retention and behaviour of aerosol materials flowing through simulated LWR plant piping without including the effect of the downstream building. The test consisted of three separate runs with different gas flow rates and CsOH:MnO aerosol ratios. These tests complement the other containment bypass tests.

Test - LA4 Late containment failure with overlapping aerosol injection periods

The objective was to perform an integral aerosol behaviour test simulating late containment failure. CsOH was injected first and this injection was overlapped with an MnO injection which continued beyond the end of the CsOH injection. Specific objectives were:

- provide a data base of aerosol behaviour under steady state conditions
- provide a link to the DEMONA tests using a steel containment and soluble and insoluble aerosols
- provide a data base for thermal-hydraulic code comparison
- determine the effect of different release timing for two aerosol species
- determine the fraction of aerosol vented during late containment failure

Test LA5 - Rapid depressurisation with a spiked pool

The objective was to study reentrainment from a flashing pool caused by rapid depressurisation which may occur during a containment failure accident. The test complements test LA6. The pool was heated to saturation temperature and was sufficiently deep to ensure bulk boiling during the depressurisation period. No external aerosol was used but the pool was spiked with a soluble and insoluble aerosol.

Test LA6 - Rapid depressurisation with aerosol injection

The objective was to study pool flashing, reentrainment and aerosol resuspension from dry surfaces during depressurisation which may occur during a containment failure accident situation. The test complements test LA5. The same soluble and insoluble aerosols used in test LA5 were spiked in the pool. Different soluble and insoluble aerosol species were used for the injected aerosol.

SCOPING EXPERIMENTS

A short series of scoping experiments (CB1,CB2,CB3) was performed before the main experiments to determine whether they were feasible. They complemented tests LA1 and LA3.

EXPERIMENTAL CONDITIONS:

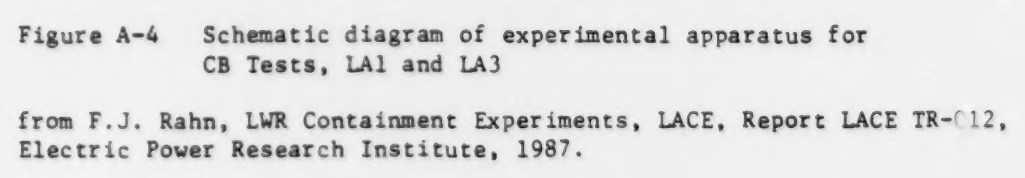
Two of the LACE tests (LA1 and LA3) focus on a containment bypass accident in a large dry PWR (called a V sequence in WASH-1400). These tests will also be relevant to aerosol deposition and resuspension in pipes under other severe accident conditions in PWRs and BWRs and also in CANDU reactors. In the V sequence it is postulated that two check valves fail in the emergency core cooling pipeline to the reactor vessel (figure A-4). Following the valve failures, overpressure in a connected low pressure pipe is assumed to cause it to rupture allowing primary system coolant water to discharge into an adjacent building, thus bypassing the reactor containment building. The loss of cooling water from this rupture eventually leads to core overheating and the release of radioactive aerosols and gases from the reactor into the pipeline. The pathway from the overheated core to the environment is tortuous so that significant deposition and/or agglomeration of aerosols may occur. Important segments of the pathway are core region, upper plenum, coolant pipes, steam generator, make up line with valves, transition sections and elbows and auxiliary building. The latter is assumed to fail by overpressure. The conditions of LA1 and LA3 together with the scoping CB tests are given in table A2.6.1.

Table A2.6.1

AEROSOL AND THERMAL-HYDRAULIC CONDITIONS AT TEST PIPE INLET
(from Report LACE TR-012, January 1987)

Test	Aerosol	Soluble Mass Fraction	Carrier Gas	Gas Velocity m/s	Temp C	Degrees Superheat	Auxiliary Building Conditions
CB1	solNaOH	1.00	Air/steam	100	186	88	saturated steam/air 85 C
CB2	solNaOH insol aluminum hydroxide	0.67	Air-steam	91	111	15	saturated steam/air 81 C
CB3	insol aluminum hydroxide	0	Air-steam	97	160	66	saturated steam/air 84 C
LA1	solCsOH insol MnO	0.42	Nitrogen- steam	96	247	141	superheated steam/air 115 C
LA3A	solCsOH insol MnO	0.18	Nitrogen- steam	75	298	208	no aux bldg
LA3B	solCsOH insol MnO	0.12	Nitrogen- steam	24	303	219	no aux bldg
LA3C	solCsOH insol MnO	0.38	Nitrogen- steam	23	300	215	no aux bldg

The containment bypass tests each used a test pipe 63mm in diameter, 27m long with five 90 degree bends, four horizontal sections and two vertical sections. A schematic is shown in figure A-4. The pressure drop across the pipe was approximately 0.1 MPa and the Reynolds number of the carrier gas approximately 500,000. The diameter of the test pipe was enlarged to 300 mm before entering an 825 cubic metre building which was vented to the atmosphere through a scrubber. For the three LA3 tests the auxiliary building was not used and the test pipe was vented to a scrubber. About 20 hours before the injection of the aerosol, steam was injected through the test pipe into the auxiliary building and was continued until steady state thermal conditions of about 83C and 1 atmosphere were reached. The auxiliary vessel then contained equal quantities of air and steam. The aerosol and carrier gas were then passed into the test pipe for 60 minutes. The aerosol characteristics for each experiment are given in table A2.6.2.



from F.J. Rahn, LWR Containment Experiments, LACE, Report LACE TR-012, Electric Power Research Institute, 1987.

from F.J. Rahn, LWR Containment Experiments, LACE, Report LACE TR-012, Electric Power Research Institute, 1987.

Table A2.6.2

MEASURED AEROSOL CHARACTERISTICS AT TEST PIPE INLET
(from Report LACE TR-012, January 1987)

Property	CB1	CB2	CB3	LA1	LA3A	LA3B	LA3C
Rate of injection g/s	3.0	0.9	2.0	1.1	0.6	0.9	0.9
Aerodynamic MMD micrometre	3.9	3.1	4.3	1.6	1.4	2.4	1.9
Geometric standard deviation	2.9	2.6	2.5	1.9	2.0	2.0	2.1
Suspended mass concentration g/cubic metre	9.5	3.2	6.1	3.6	6.9	22.0	30.2

The results of the tests can be summarised quite briefly. In the CB tests, whether the aerosol was a liquid or a dry solid, very little of the material was retained in the test pipe. The proportion of the solid aerosol retained (CB3) was lower than the liquid aerosol both in the 63mm and 300mm pipes. A larger fraction of the solid aerosol was suspended in the atmosphere of the auxiliary building. This suggests, as would be expected, that the physical nature of the aerosol has an important effect on its behaviour in pipe flow, possibly because a dry solid aerosol is more easily resuspended.

The aerosol/gas stream entered the auxiliary building at high velocity (200m/s) horizontally near the bottom of the building. A large fraction of the aerosol mass leaving the test pipe impinged on the building wall and floor and was not suspended in the atmosphere. For CB1 and 2 the impinged fraction was approximately 80% and for CB3 40%. The fraction of the aerosol available for release to the environment would thus be significantly reduced.

The major results from the CB tests were:

- Aerosol retention in 63mm pipe less than 5%
- Aerosol vented from auxiliary building about 2% for CB1 and CB2 and 20% for CB3
- Test pipe removed about 90% of liquid aerosol in CB1 and CB2 and 50% of solid aerosol in CB3
- About 20% of entering aerosol mass was suspended

in the auxiliary building in CB1 and CB2 and 60% in CB3

- Physical state of aerosol important to its pipe flow behaviour and fraction suspended at the outlet
- In the test pipe a large fraction of the entering aerosol deposited and then resuspended or flowed as a liquid film along the pipe
- A large fraction of the aerosol entering the auxiliary building impinged on the wall or fell out on the floor
- The characteristics of aerosol passing through piping at high velocity are changed significantly resulting in an enhanced deposition in the auxiliary building

LA1 and-LA3 results

The differences between the CB tests and the LA1 and LA3 tests were:

- more realistic aerosol (CsOH and MnO)
- higher carrier gas temperature (280C)
- nitrogen as non condensible carrier gas
- measured aerosol flow distribution
- improved in situ trapping of deposited aerosol
- superheated steam/N atmosphere (115C) in aux bldg
- improved measurement of aux bldg steam fraction
- additional wall condensate collectors in aux bldg

In test LA1 virtually all of the aerosol material (>98%) was retained in the 63mm test pipe. The reason appears to be the physical nature of the aerosol which formed a viscous, tightly adhering film on the pipe surface.

In test LA3 the configuration was similar to LA1 except that the auxiliary building was not included. the aerosol composition and flow were varied in the two tests by factors of 5 and 4 respectively. More than 70% of the injected aerosol, whether soluble or insoluble, was retained in the 63mm test pipe, with the exception of test LA3B when more than 40% of the soluble aerosol was retained.

LA2 results

The objective of test LA2 was to simulate a postulated accident where a failure to isolate containment occurred. In addition to measuring the quantity and particle size spectrum of the leaked aerosol it was intended to determine the effect of leak path location i.e. whether at the top or bottom of the containment vessel. This might be of major importance if stratification of the aerosol should occur. The leaks were 18mm diameter orifices situated 16.5 and 4 m above the containment vessel bottom.

The system was preheated to achieve steady state conditions and the CsOH and MnO aerosols injected directly into the containment for 50 minutes during which time the leak paths were open. 98% of the aerosol mass was recovered and the integral fractions of settled, plated and leaked aerosol mass were 0.501, 0.114 and 0.385 respectively. No significant difference was found in the mass of aerosol leaked through the upper and lower leak paths. About 64% of the aerosol entering the leak paths was deposited in the duct before entering the scrubber. This is a decontamination factor of 2.8 and shows that even a simple leak path (orifice plate in duct) can cause a significant reduction in the mass leaked. The major process involved is believed to be turbulent deposition. The initial particle size was an AMMD of 1.3 micrometre and this increased steadily to a maximum of 8 micrometres after 200 minutes and then decreased rapidly. The geometric standard deviation fluctuated between 1.6 and 2.1 during the same period. This particle growth by agglomeration and water condensation is believed to have enhanced deposition rates.

LA4 results

The objective of this test was to determine aerosol behaviour with overlapping injection periods and a late containment failure. The experimental configuration was similar to LA2 except that the lower leak path was removed. The pressure in the containment rose to 3 bar and was maintained constant for 200 mins, after which the containment was vented. No detailed results are given for Test LA4 in the report.

THERMAL-HYDRAULIC COMPUTER CODE COMPARISON

The results of an intercomparison on various thermal-hydraulic computer codes is given in figure A-5. The participants were given the actual thermal-hydraulic input data (steam flow rate and fraction, gas temperatures etc) and asked to compute the final conditions (total pressure, condensation rates etc). After the calculations were complete they were compared with each other and the test data. As figure A-5 shows, some of the agreement was rather good.

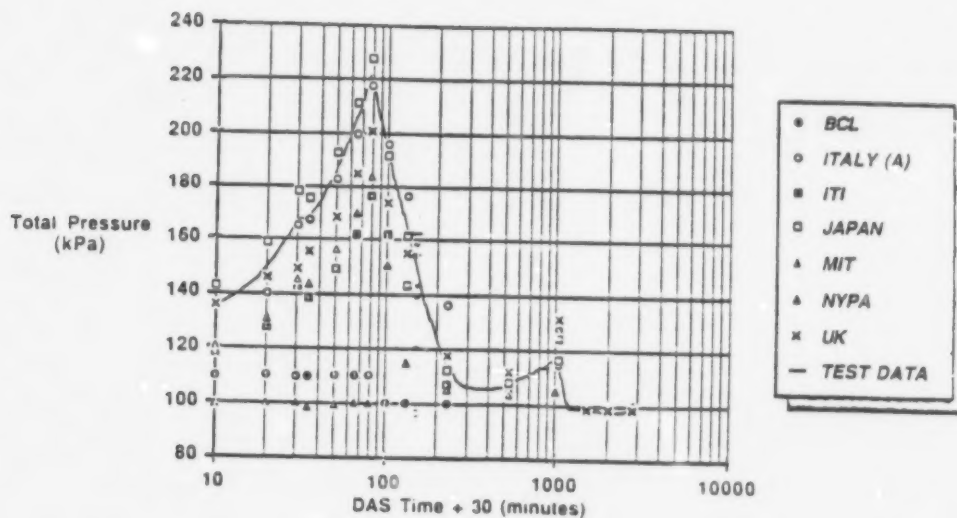


Figure A-5 An example of the blind post test comparison done using thermal-hydraulic codes for test LA2. Many of the codes gave quite good results, while others gave poorer results, indicating a deficiency of the code models or the inputs to the code.

from F.J. Rahn, LWR Containment Experiments, LACE, LACE TR-012, Electric Power Research Institute, 1987

A 2.7 DEMONA EXPERIMENTS TO INVESTIGATE AEROSOL BEHAVIOUR WITHIN A CONTAINMENT

(Abstracted from Kanzleiter et al IAEA SM 281/43, 1986)

The purpose of the experiment was to demonstrate experimentally the aerosol retention capacity of a large dry PWR containment for core melt down accident sequences which involve late overpressure failure of the containment. A second aim was to demonstrate the validity of the aerosol and thermodynamic codes such as NAUA, COCMEL and FIPLOC for the prediction of core meltdown accident sequences. For the DEMONA experiments, the Batelle Frankfurt model containment (640 cubic metres) was equipped with steam and aerosol generators and appropriate thermal and aerosol instrumentation. At the time of the report, six of the ten planned main experiments had been completed with tin oxide and silver aerosols at 12 g/cubic metre concentration (maximum) in air or steam-air atmospheres using a quasi-single compartment containment configuration. The experimental results to date have revealed a significant dependence of the aerosol behaviour on the thermodynamic conditions of the containment atmosphere. Aerosol depletion develops three or four times faster in a condensing steam-air atmosphere than in a dry air atmosphere due to condensation and enhanced gravitational settling.

NAUA code :

The NAUA code predicts aerosol removal from the atmosphere of an LWR containment during a core melt down accident. It is based on a detailed description of all the micro-scale physical aerosol processes involved such as agglomeration, steam condensation on and evaporation from particles, sedimentation, diffusion, thermophoretic and diffusio-phoretic deposition and inertial or turbulent deposition. The aerosol parameters involved in the description of these processes were derived from separate-effect tests.

COCMEL code

NAUA calculations need thermodynamic input and an extended version of the COCMEL code was derived for this including a special module for computing the condensation rates in a containment atmosphere. COCMEL and NAUA are single node codes calculating the average values for a compartmentalised containment.

FIPLOC code

FIPLOC is a multimode code coupling detailed thermodynamic modelling with rather simple aerosol models. It is still under development and is based on the RALOC code.

EXPERIMENTAL PROCEDURE:

The model containment is built from conventional reinforced concrete without a steel liner and is therefore not leaktight. Under DEMONA operating conditions (3bar, 115C) the leak rate is less than 3%/h. The aerosol generator produced a metal oxide aerosol of primary particle size about 0.2 micrometre and could provide a concentration of 12g/cubic metre within less than one hour. The experimental procedure is illustrated in figure A-6. Air is first expelled from the containment by steam injection to a pressure level of 1 bar. The containment is then heated by injecting steam to a pressure of 1.7 bar and a temperature of 115C over a period of two days. Hot air is then injected with the suspended metal oxide until, after about 1 hour, the specified conditions of 3 bar and 115C are reached. The aerosol is then depleted under steady state thermal conditions (controlled continuous steam injection). This phase lasts from 24 to 36 hours.

RESULTS:

A comparison of experimental results and those calculated from NAUA is given in figure A-7. At the end of the one hour injection period only about one third of the injected aerosol was still airborne. After one and a half hours the aerosol concentration had decreased by an order of magnitude. After 36 hours the concentration had decreased by a further 3 orders of magnitude. The initial aerosol was about 0.2 micrometres diameter and, when the concentration was high, most of the primary particles coagulated to form larger flaky aggregates which sedimented out rather rapidly. Only one per cent of the aerosol plated out on walls and ceiling, confirming that the main removal process was sedimentation. The dry results are compared with those from wet experiments in figure A-8. Aerosol removal is obviously much enhanced in the wet probably due to diffusiophoretic effects and condensation on particles. Aerosol removal in wet conditions is about three to four times faster than in dry. From the results available so far it is apparent:

- (i) That the results are reproducible
- (ii) The results are consistent
- (iii) High aerosol concentrations (>1g/cubic metre) lead to a high depletion rate almost independent of the thermodynamic conditions while lower concentrations result in lower depletion rates.
- (iv) Intensive steam condensation enhances aerosol removal significantly.

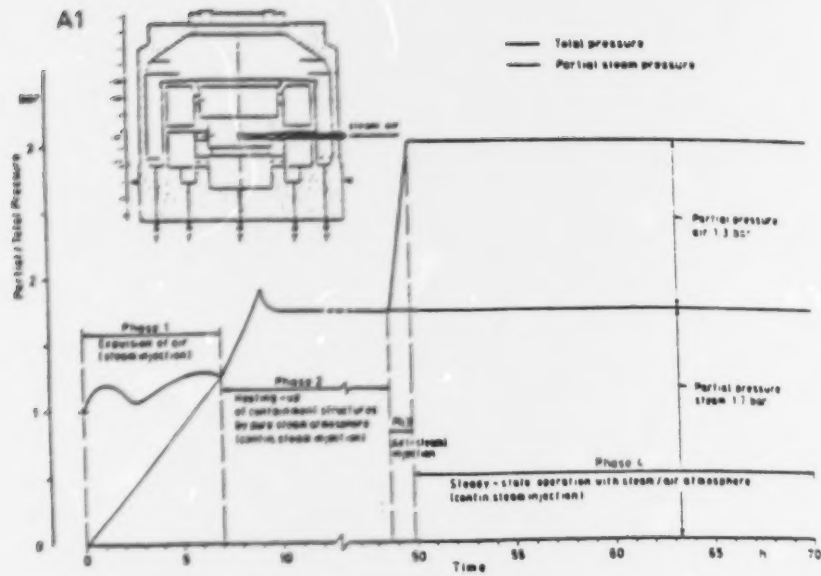


Figure A-6 Experimental arrangement for DEMONA experiment from Kanzleiter et al, IAEA-SM-231/43, 1986

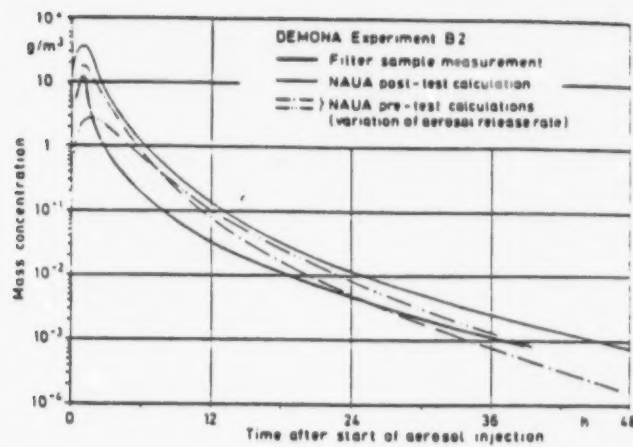


Figure A-7 Comparison of measured and calculated aerosol mass concentration in DEMONA dry experiment B2 from Kanzleiter et al, IAEA-SM-281/43, 1986

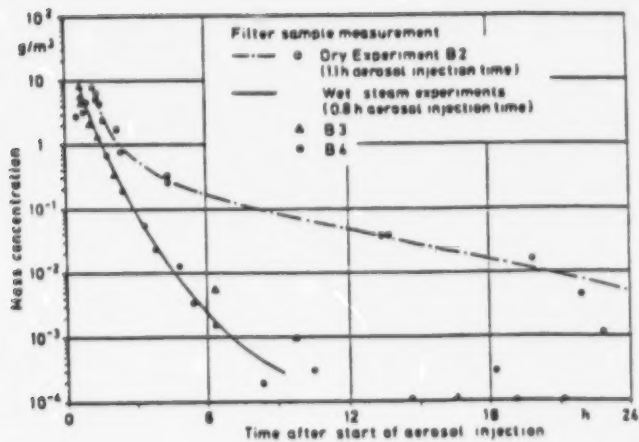


Figure A-8 DEMONA experiments: comparison of measured aerosol mass concentration in dry experiment B2 and wet experiments B3 and B4 from Kanzleiter et al. IAEA-SM-281/43, 1966

A 2.8 CSNI Task Force on Source Terms (Second Draft)

TASK VII FISSION PRODUCT/AEROSOL TRANSPORT IN CONTAINMENT AND

OTHER BUILDINGS. Abbey and Dunbar undated and without references

This paper reviews (mainly US) studies and concerns itself mainly with:

1. Inside buildings - steam condensation/evaporation on aerosols
 - aerosol agglomeration
 - aerosol deposition on floors and surfaces
 - resuspension of deposited material as a result of steam explosions, hydrogen combustion or rapid depressurisation
2. Within leak paths in building walls
 - aerosol agglomeration
 - aerosol deposition
 - aerosol resuspension

Superimposed on all these is the effect of radioactive decay and possible self heating as a result. Probably a second order effect.

Removal processes:

Modelling is complicated due to growth and evaporation of particles. The removal rate as a function of radius has a pronounced minimum between 0.2 and 2 micrometres. Brownian diffusion causes rapid removal of smaller particles but for dense aerosols agglomeration is rapid and shifts the particle size upwards, so diffusion relatively unimportant. In most containment applications removal rate dominated by how fast particles grow to a size at which gravitational settling becomes rapid. Picture may be modified when there are substantial heat or mass flows to the walls but in LWRs thermal gradients are usually small at the walls because heat can be transported as latent heat with the condensing steam. For diffusiophoresis to be important significant steam condensation must coincide with aerosol presence in the containment.

Growth processes:

If mean radii are between 0.1 and 1 micrometre and mass concentration is greater than 1g/cubic metre, agglomeration is an important mechanism of particle growth. However there is also a dip in the agglomeration rate between 0.2 and 2 micrometre. for a size distribution starting near its most stable position it typically takes about 2 h or less for a significant proportion of the particles to agglomerate to sizes where settling becomes rapid on the time scale of an hour.

If conditions are such as to allow steam condensation, growth is much faster, however very sensitive to the thermal hydraulics.

Gravity based processes are slower but more reliable and are largely independent of thermal hydraulics.

ASSUMPTIONS IN EARLY CODES:

1. Containment well mixed (i.e. number concentration the same everywhere except in the boundary layers next to surfaces)
2. Radionuclides well mixed over the size distribution
3. Once aerosol is deposited it cannot become airborne again.

Codes are based on several well mixed volumes with transport between them have been developed. Codes have also been developed to follow the distribution of the components separately over the size range. It is difficult to model resuspension but it is important to try since this is the major barrier to claiming substantial containment retention in late failure sequences.

AEROSOL MODELLING:

There is wide agreement between various codes both on the individual models and on the assumption that the rates for the different processes can simply be added. The APS study only considers diffusiophoresis from method of steam condensation gradients and doesn't consider the more recent mass flux method, recognises the need for validation of gravitational and turbulent agglomeration and calls for small scale separate effects experiments to validate models.

APS and ANS studies describe the model in the TRAP-MELT code which is based on mass transfer alone. Recent calculations have shown that heat transfer effects have a major role in deciding whether vapours condense onto particles or walls and therefore TRAP-MELT is unreliable. It also does not take account of the addition of mass to the aerosol particle which, in the containment, is precisely what is of interest.

WEAKNESSES IN METHODOLOGY:

1. Documentation of methods - generally not enough detail given to make a proper scientific peer review.
2. Information on shape factors and effective densities - SANDIA QUEST study identifies these as among the most sensitive parameters in modelling of aerosol behaviour.
3. Experimental test of gravitational and turbulent agglomeration
4. Hygroscopicity effects - information on the water vapour pressures above the hygroscopic components of the aerosol is

required together with the modelling for including such effects in the codes.

5. Thermal-hydraulic calculations and experiments on intra-cell flows

6. Experimental and theoretical information on the flashing of pools and possible resuspension of aerosols during sudden depressurisation and also on steam condensation on aerosols under these conditions.

7. Resuspension as a result of hydrogen combustion or steam spikes.

8. Size distribution of water droplets during blow down - what fraction of the coolant remains as a persistent aerosol.

9. Retention in leak paths, also identifying leak paths.

A 2.9 LEAKAGE OF AEROSOLS FROM CONTAINMENT BUILDINGS:

H.A. Morewitz Health Physics 42,2, Feb 1982

Experimental data on the plugging of ducts by aerosols are reviewed and compared with a simple model. The model predicts that the mass of aerosol entering the duct prior to plugging is proportional to the cube of the duct diameter. This relation has been used to correlate experimental data of a variety of aerosols over a range of duct diameters from 100 micrometres to 30 cm. The proportionality constant appears to be a function of the effective density of the aerosol deposit and of the location of the plug. In the process of plugging leak paths, aerosols attach to the walls or to previously deposited aerosols. Some of these agglomerates break off and are resuspended in the airstream so that the sizes of aerosols exiting from short length leaks are substantially increased. When all of these effects are considered it appears that < 1mg of respirable aerosol can leak from a typical reactor containment building operating at the design pressure and leak rate (0.1% vol/day). This is a reduction of more than four orders of magnitude from the case where the aerosol is assumed to leak as a gas.

Starts with anecdotal history showing that aerosols do not pass through leak paths in structures as if they were gases, but, given high enough concentrations and long enough times they plug the leak paths. e.g. gas leakage from pressurised cabins in passenger aircraft decrease with time due to plugging of leak paths by cigarette smoke. Many experiments are quoted showing attenuation of aerosol on passing through leak paths. (Witherspoon, 1970 and Witherspoon and Postma, 1971)

Hilliard et al (1980) studied the plugging of ducts of 2.21 to 25.5 cm inside diameter with aerosols of sodium carbonate and partially hydrated NaOH. They showed that the mass of aerosol entering the duct prior to plugging was proportional to the duct diameter and that the proportionality constant was 40g/cc.

Data show that when 10-70 mg of aerosol has entered a 1mm dia capillary it will plug (such a capillary will leak gas at the entire design leak rate of 0.1%vol per day from a RCB). Furthermore the experiments indicate that the size distribution of exiting aerosols is shifted to larger sizes by agglomeration within the capillary so that < 1mg is transmitted as respirable aerosol even through a short length 1mm dia capillary. However real leak paths through the concrete walls of RCBs are long irregular cracks which have rough surfaces so that additional aerosol removal phenomena, such as impaction at bends, are operative and reduce the transmitted aerosol mass still further. Thus the mass of respirable aerosol leaked at the design pressure from an intact RCB will be < 1mg total rather than the 10 to 600 g per day which will leak if the aerosol is assumed to leak as a gas.

A 2.10 FACTORS AFFECTING PRIMARY SYSTEM RADIONUCLIDE RETENTION IN AN LWR SEVERE ACCIDENT by A.T.D. Butland and M.R. Kuhlman
abstracted from IAEA SM-281/20, 1986

This paper discusses two postulated PWR and two LWR accidents. PWR accidents are (WASH 1400) S2D and TMLB'. Former involves a small primary circuit break (9.5-50mm equivalent diameter) and the failure of the high head ECCS pumps. Primary water boils off through the break until the core uncovers, overheats and melts, releasing fission products to the primary system gas space.

TMLB' is initiated by a transient event accompanied by the loss of on-site and off-site power, so that the secondary side feed water pumps and ECCS become inoperative. Primary coolant heats up and boils off through the Power Operated Relief Valve (surely pressure operated relief valve - there is no power!), eventually leading to core uncovering and melt at full circuit pressure (why full circuit pressure if coolant has boiled off?). Flowpath from the core to the containment therefore via the vessel upper plenum, the hot leg, the pressuriser and the relief tank.

The paper calculates gas and surface temperatures for the four accident sequences (gas temperatures up to 2500 C at the core exit for the PWR and up to 1760 C for the BWR), the fraction of fission products released for the core (Xe,Kr,I,Cs .75 and .952 for the PWR accidents and 1 for the BWR accident). Inventories of fission products given in Kg (PWR, Xe,Kr,I and Cs 319,23.5,12.0, 167.4; BWR 412,27.3,17.7, 220). Composition of coolant gas deduced for each case. Point made that presence of hydrogen (for the S2D sequence can reduce the density and viscosity of the gas by factors of 6 and 2 respectively, thus altering the transport of any aerosol released and affecting aerosol retention calculations by up to 50%.

It is assumed in all primary system material transport codes that the released noble gases and vapours will be transported with the steam and hydrogen gas stream and will be at the temperature of the gas. At the PWR temperatures CsOH and CsI will be mainly gaseous until the steam generator is reached but silver and silver telluride if present would begin condensing out and forming aerosols in the core and upper plenum. This is important because most computer codes do not model silver telluride and calculate substantial tellurium retention by chemisorption on primary system surfaces.

Other released materials, such as Fe and In, would condense much sooner and would probably exist as aerosols at the core exit and it is reasonable to assume that the released mass of these materials exists as aerosol at the core exit, which is done by most computer codes like TRAPMELT2. Situation less clear with Ba, Sr and Ru because of uncertainty over the chemical species. It may not be correct to assume that they are released from the core as aerosols. Cadmium clearly released as a vapour (BP 765 C)

and may react with other species in the circuit - not correct to assume it is released as an aerosol unless it reacts soon after release to form a condensible species.

THERE IS THEREFORE A NEED TO MODEL HOMOGENEOUS NUCLEATION AS WELL AS CHEMICAL SPECIES FORMATION ALONG THE TRANSPORT PATH. This can be done with the RAFT code (Im et al, 1984), but not with TRAPMELT or RETAIN. However this may not be very important since there will probably be SOME aerosol released to provide condensation centres for heterogeneous nucleation with which TRAPMELT2 and RETAIN can deal. This of course depends on the particle size of the aerosol. TRAPMELT assumes a number mean particle radius of 0.05 micrometres and a geometric standard deviation of 1.7 (based on experimental data from vapourising and condensing fuel).

Aerosol agglomeration processes:

Four agglomeration mechanisms modelled by most codes - Brownian, gravitational, and two forms of turbulent agglomeration. All codes use the Schmoluchowski (see Zebel, 1966) expression for Brownian coagulation. Most codes use an expression for gravitational agglomeration involving terminal velocities of particles involved and a collisional efficiency, e , using Pruppacher and Klett's (1978) expression for e , since it takes proper account of the relative particle motions.

Turbulent agglomeration arises in two ways - variations in the gas turbulence cause collisions due to the motion of the aerosol particles within the gas (turbulent shear agglomeration); differences in particle inertia cause turbulent inertial agglomeration. Most codes use an expression for these given by Saffman and Turner (1956) but experimental validation needed for the multiplicative constants at the high number densities here.

Calculations with TRAPMELT2 show that for particles < 1 micrometre Brownian mechanism is predominant, for larger particles gravitational deposition is more significant unless the particles are of equal size when turbulent agglomeration is the most important.

The combined effect of the four mechanisms is calculated by adding the two turbulent and the gravitational components in quadrature and then separately adding the Brownian component. Not clear whether this is correct but it may not matter because one component usually dominates. Main problem is that the formulae have not been verified for the high particle number densities predicted in the primary system.

Chemical interaction between vapours and aerosols

Particle concentration of 5×10^{13} per cubic metre are possible and surface area of about 600m^2 per cubic metre - much larger than area of primary system structures and so chemical interaction

between vapours and aerosols might be very important, e.g. an interaction between Te and Ag to form silver telluride would reduce tellurium retention in the primary system. These interactions not modelled in TRAPMELT2 nor in any other primary system aerosol codes.

Well mixed assumption

It is usual to calculate transport and retention by dividing primary system into well mixed control volumes with uni-directional flow between them. This is true for TRAPMELT2, RETAIN and RAFT. New developments adopt a Lagrangian approach by which a plug of gas is followed through the primary system. Each has advantages and disadvantages. Lagrangian approach seems natural for a pipe but is difficult to justify in a large volume where recirculation could occur.

Deposition, resuspension and relocation

Vapour deposition of volatile fission products

Most current treatments concentrate on CsI, CsOH and Te since these were believed to be the main radiologically significant volatile species. Vapours may condense on RCS surfaces, on suspended particulates, and/or react chemically with surfaces or particles. Condensation on surfaces modelled in a fairly straightforward way in the codes. Vapour pressure of the species of interest is compared with the partial pressure at the surface and allowed to approach equilibrium. Rate at which condensation will occur limited by the mass transfer coefficient computed by the code for the system of interest. This process competes with condensation on particles (when this is possible), reaction with surfaces and transport out of the volume of interest.

The key parameter governing deposition of volatiles is the identity of the species. Condensation on particles governed by temperature of the gas, concentration of the vapour and availability of particle surface. These are all dynamic quantities and thus the timing of the vapour release relative to the particle release is of great importance. Large amounts of the Cs-containing species are released early in the meltdown period, prior to the release of particles from the fuel and structures. Depending on the amount of aerosol released from the control rods, this mechanism of retention of the volatiles may or may not be important in the region where the gas is sufficiently cool for condensation. In PWR analyses using TRAPMELT, the condensation of Cs containing vapours on particles is frequently the major route for RCS retention of these species. In the absence of high concentrations of control rod aerosol early in the melt process this would not be the case.

The extent of surface condensation of vapours will be determined by the temperature of the RCS surfaces, the gas concentration of the volatile species and the retention time of the vapour in the

area where condensation is possible. This process competes and is therefore closely coupled with condensation on particles, so that surface condensation depends on the concentration of aerosol. In the analysis of the above accidents condensation on particles dominates surface condensation by factors of 1.4:1 to 7:1, depending on the sequence. The high pressure sequences with their longer residence times and greater aerosol concentrations have the higher ratios.

Particle deposition

The settling of particles at $Re < 1$ well described by the Stokes terminal velocity which is proportional to the square of the particle diameter, its density and inversely to the gas viscosity. For larger particles the settling velocity is modified but this doesn't matter much since they deposit out so rapidly anyway. Range of particle densities anticipated for the RCS aerosol is 3.5 - 10 g/cubic centimetre. Initial particle diameters for the involatile materials released from the core are estimated to be around 0.05 micrometre but particles of around 10 micrometre are possible when high concentrations experience long residence times. The latter are not transported efficiently through the RCS.

The RCS geometry, particularly the ratio of horizontal area to the volume of the container is important in determining rate of aerosol sedimentation, but is not easy to quantify. Effective settling area in any vessel with internal structures not easy to determine due to possible flow channelling or recirculatory flows which could increase or decrease the effective area. These factors are not taken into account in current analyses and it is not easy to see how to do so.

In most cases gravitational settlement is the most effective mechanism for aerosol retention, especially when there is a prolonged period with very low flow rates during which agglomeration may cause particle diameters to increase above 1 micrometre. More important for high concentrations, therefore must understand control rod aerosol release.

Turbulent deposition

Turbulent deposition is most effective in very high flow rates, deposition velocity depends on particle relaxation time and fluid velocity. Most important in small diameter pipes (high Re) and long pipes. Present studies indicate that only under the extreme conditions of a V sequence (15 cm dia pipe, flows > 300 m/s and pipes > 60 m long) will it contribute significantly to total retention, but these don't take resuspension into account and for dry deposits this may be substantial.

Diffusive deposition

This is inversely proportional to particle diameter and fluid viscosity. Influence greatest for small particles and not a great contributor to the mass of material retained.

Thermophoretic deposition

This is difficult to quantify as one needs to know the thermal gradient and hence the boundary layer thickness. An experimental study and comparison of code predictions with the results is underway in the USA (no reference).

Impaction and deposition on obstructions

Most analyses treat the RCS as a set of connected well-mixed volumes. Inertial deposition, other than sedimentation, is not treated rigorously in existing codes despite the tortuous paths and multiple obstacles that the flow encounters in the RCS; treatments that do exist simply consider impaction at bends. Better definition of flow fields and geometry is required. Until this is done particle retention may be underestimated.

Resuspension and relocation

Fission products deposited by condensation may be resuspended if the cool surface on which they have condensed becomes heated later in the accident either by more core melting or by decay heat. Increases in flow may also resuspend particles. Current experimental work at ORNL and an analytical treatment at CEGB, Berkeley. Liquid deposits may also move by gravity or gas flow. Is being studied in the LACE programme.

OTHER PHENOMENA WHICH MAY AFFECT AEROSOLS

Radioactive decay

The two decay chains:

Te131 25m.....I131.....8d.....Xe131 stable

Te132..... 78h.....I132.....2.3h....Xe132 stable

may affect the quantity of iodine transported around the reactor circuits, for example tellurium deposited on a primary system surface could re-evolve as water soluble iodine.

Chemical change

If boric acid is present in the coolant it could break down CsI into complex caesium borates and HI. CsOH can also react with boric acid to form borates. (Bowsher et al, 1986)

Radiolysis

All the chemical data used in computer codes are based on experiments in a radiation-free environment. An intense radiation field may affect chemical reactions significantly, e.g. CsI reacts more strongly with steel surfaces in the presence of radiation forming caesium compounds and releasing iodine. Work on this in progress at the Sandia Laboratories.

Photophoresis

This is the result of a temperature gradient IN THE PARTICLE causing different momentum changes to the gas molecules hitting the particles on opposite sides and thus affecting the particle movement. Temperature gradients about 100 times greater than in the gas are required for photophoresis to be comparable in effect to thermophoresis. It may cause accelerated drift from a hot source (the core) to reactor structures but is unlikely to be important compared to convection and turbulence.

CONCLUSIONS

1. A number of different species can be released and the presence of large aerosol concentrations will affect transport and deposition. More experiments and computer modelling are needed.
2. Large aerosol particle number densities may cast doubt on agglomeration models now used in computer codes.
3. There has recently been a move away from the well mixed assumption to codes based on Lagrangian approach. This seems worthwhile although recirculation and turbulence must not be neglected.
4. The retention of volatile materials by condensation is important and accurate estimates require better knowledge of RCS temperatures and relative magnitudes of aerosol and RCS surface areas than is available. Inadequate knowledge of circulation patterns in vessel hampers accurate aerosol deposition estimates. Better knowledge of available settling area and modelling of turbulent deposition is needed.

A 2.11A CHEMICAL EQUILIBRIUM ESTIMATE OF THE AEROSOLS PRODUCED IN AN OVERHEATED REACTOR CORE

by R.P. Wichner and R.D. Spence Nuclear Technology 70, 376-393
Sept 1985

The degree of vapourisation of light water reactor core materials estimated using an idealised procedure involving (a) specification of the phases that are present for both structural and fuel material (b) estimation of the vapour pressures exerted by the individual components of each phase (c) assuming a degree of vapourisation of each phase constituent, allowing equilibration between gaseous and condensed species within the assumed pressure vessel volume.

For PWRs the aerosol was estimated to consist mainly of silver, indium oxide, caesium hydroxide and cadmium and for BWRs caesium hydroxide, caesium iodide and tellurium. If boron is present it would alter significantly or dominate the composition of the aerosol. The structural materials make up <9% of the aerosol (36 to 57Kg), as compared with 17kg from severe accident sequence analysis studies and 10.7Kg (Parker). SASCHA data used in NUREG-0772 give much higher estimates at 295 and 250kg.

In spite of its idealised assumptions this is an impressively comprehensive paper and the agreement with other estimates is good. It is assumed that the core is brought to a temperature of 2700 K. This is above the melting point of the pressure vessel but the molten mix and the associated vapours are assumed to be contained within the pressure vessel.

A 3.1 AEROSIM

S.A.Ramsdale AEROSIM Input and Output, UKAEA Report: NST/84/245, 1984 (This synopsis has been abstracted from Nuclear Aerosols in Reactor Safety, supplementary report, Nuclear Energy Agency, OECD, Paris, 1985)

AEROSIM is an aerosol agglomeration and deposition code which calculates the time development of arbitrary particle mass distributions using a finite elements method. Originally designed for LMFBR accidents, it has been adapted for PWR use by the inclusion of a model for diffusiophoretic deposition on the walls.

Programming strategy:

The particle number density distributions over the continuous range of particle masses is approximated by dividing up the range of masses into a number of finite mass intervals and assigning to each interval a number density. The integro-differential equation governing the behaviour of the original distribution is replaced by a set of coupled non-linear ordinary differential equations. The distribution is thus not constrained to be lognormal; any distribution consistent with the resolution of the discretisation can develop.

Input:

The main classes of input variable are:

- building properties
- properties of the carrier gas
- properties of the aerosol material
- aerosol behaviour correction factors
- aerosol distribution at zero time
- aerosol source rate and size distribution
- diffusiophoretic removal rate and duration of operation
- range of particle masses and number of intervals
- times at which output is required.

Calculation of coefficients:

The input is read into a pre-program AERODAT, which then constructs a data file for the main program AEROSIM. In particular AERODAT calculates the coefficients for the equations: source, removal and agglomeration rates, as well as the initial values. This strategy severely limits the extent to which these coefficients can vary with time.

The reason for this is that the coefficients for the discretised equations are not calculated by evaluating the coefficients of the continuous equations at values of a discrete set of sizes. Instead they are associated with size intervals and are suitably

weighted integrals (double integrals in the case of agglomeration coefficients) of the original coefficients over those intervals. Because the calculation of these integrals is time consuming they are evaluated once only, at the beginning of a run.

Solution of the equations:

The equations constructed in AERODAT are integrated in AEROSIM using the HARWELL developed program FACSIMILE. This uses a Gear's method of solution. There is automatic adjustment of the time step length during the integration.

Output:

At each of the times selected by the user the variables are printed out. There are two types of variable: masses of material removed by diffusiophoresis, by other removal processes and by leakage from the building, and properties of the material still airborne.

Modelling of physical processes:

The following processes are modelled by AEROSIM and can therefore be included provided the necessary output is available:

- Brownian agglomeration
- gravitational agglomeration
- turbulent agglomeration
- removal by gravitational sedimentation
- removal by diffusion to surfaces
- removal by thermophoresis
- removal by diffusiophoresis

The initial and source distributions are calculated in AEROSIM using the lognormal formula, but this can be bypassed and arbitrary distributions can be put directly into AEROSIM.

The following processes are NOT modelled in AEROSIM:

- removal by turbulent deposition
- condensation on and evaporation from the particles
- electrostatic effects
- resuspension (except where modelled in the source term)

The modelling is restricted to aerosol in a single well mixed compartment. Because agglomeration and removal coefficients are calculated once only, before the start of the integration, the properties of the carrier gas have to be introduced as time averages. The source rate can be varied with time but the source size distribution is constant.

Of the possible steam related phenomena, only diffusiophoretic removal at surfaces has been included in AEROSIM. The program

is therefore applicable to the case where the greater part of steam condensation is on surfaces rather than on aerosol particles.

Status of code:

A developed version of the original LMFBR code is available. The numerical methods employed have been verified and the predictions of the code have been shown to match exact analytical solutions for a wide range of input data. comparisons with some large scale integral experiments have also given satisfactory results. The use of AEROSIM for PWR accidents has not been validated; in this context it should be regarded as being at an early stage of development.

A 3.2 AEROSOLS

l'Homme et al Presentation du Code AEROSOLS/B1, CEA Report, November 1983 (This synopsis abstracted from Nuclear Aerosols in Reactor Safety, supplementary report, Nuclear Energy Agency, OECD, Paris, 1985.

Three versions have been developed to date; A1 for study of sodium fires in LMFBR accidents, A2 an extended version relevant to PWR accidents, (both handle the time development of the particle mass distribution by the method of moments), B1 a version which replaces this assumption with a finite element technique and is applicable to LMFBR and PWR accidents.

Programming strategy:

A1 follows the evolution of the zeroth, first and second moments and reconstructs the distribution assuming it to be log normal. The finite element method followed in B1 takes as its variable particle numbers, one associated with each of a finite set of masses (up to 101 mass nodes are allowed). The containment can be compartmentalised into zones (up to 20 in A1 and A2, up to 5 in B1). There are then separate sets of equations for each zone, with coupling terms representing inter-zone flow.

Input:

Input for A1 falls under the following headings:

- number of zones and zone geometries
- properties of aerosol material
- aerosol behaviour correction factors
- initial aerosol distributions
- numbers of time points for the time tables

The following input is in the form of time tables, each with a list of times and the corresponding values of the variable under consideration:

- specified times of obligatory output
- aerosol source distribution (zone 1 only) at specified times
- pressure in containment at specified times
- temperatures in each zone at specified times
- temperature gradients at walls in each zone at specified times
- diffusional boundary layer thicknesses in each zone at specified times
- leak rates from each zone at specified times
- flow rates between pairs of zones at specified times

In A2 and B1 the following additional input is required:

- thermal conductivity of aerosols
- molar mass of aerosols

- van't Hoff's coefficient
- atmosphere composition at specified times.

Calculation of Coefficients:

The coefficients are calculated at each time step, the time dependence of the containment properties being given by linear interpolation of the data from the input time tables. In A1 the containment atmosphere is taken to be air, but in A2 and B1 it is a specified mixture of carbon monoxide, carbon dioxide, hydrogen, water and oxygen.

Subroutines are incorporated which calculate the physical properties of such mixtures and describe the steam-water equilibrium.

Solution of the equations:

In A1 the equations are solved using the Runge-Kutta and Adams-Moulton fourth order integration methods. A2 and B1 use the STEP routine developed at Sandia.

STEP uses a modified divided different form of the ADAMS PECE formulae. Local extrapolation is used to improve absolute stability and accuracy.

The code adjusts its order (maximum order = 12) and step size to control the local error per unit step in a generalised sense. Special devices are included to control round off error and to detect when the user is demanding too much accuracy.

Output:

Intermediate results are stored at time steps satisfying at least one of the following criteria:

- first and last time steps
- discontinuity in input data
- aerosol concentration has extremum with respect to time
- rate of change of concentration exceeds specified rate
- time since last output exceeds specified value

These values are subject to further calculations which produce as output the following variables for each zone and again for the whole containment:

- mass balance
- fractions of input mass still airborne, settled, deposited and leaked
- aerosol concentrations still airborne, settled, deposited and leaked

- aerosol size distributions
- rates of aerosol processes

In addition to this A1 output, A2 has:

- mass of condensed water on aerosols
- atmospheric percentage saturation

These calculated variables are stored as synthetic tables which are then printed out when one of the following conditions is met:

- after 50 time steps obeying the above criteria
- final time is reached
- time limit is about to be reached

Modelling of physical processes:

The following phenomena are modelled in the code:

- aerosol source, instantaneous or function of time
- gravitational settling (Stokes Law)
- deposition at walls due to Brownian diffusion
- deposition at walls due to thermophoresis (Brock's Law)
- Brownian, gravitational and turbulent agglomeration
- leakage
- inter-zone flow

The usual correction factors are taken into account, as follows:

- density correction factor
- Cunningham's slip correction factor
- dynamic shape factor
- gravitational collision efficiency

The condensation of steam on particles is modelled using Mason's equation.

Status:

All the codes are written in Fortran IV. A2 has been used to provide predictions for MARVIKEN experiments and it is still under development. B1 is still under test.

A 3.3 HAA-4A

J.M. Otter and E.U. Vaughan, HAA-4 Code Description and User's Manual, Rockwell International Report, AI-DOE-13528, Sept 1985. This synopsis abstracted from Nuclear Aerosols in Reactor Safety, Nuclear Energy Agency, OECD, Paris 1985

HAA-4A is a computer code for calculating fission product transport and removal in control volumes. Original development and use was for LMFBR accident analysis but it has been adapted and used for LWR analysis by including vapour removal models. Aerosol behaviour equations are solved using the moments method, assuming that the particle size distribution is log normal. Separate accounting is done for three components assuming particle size independent composition.

Programming strategy:

The integro-differential aerosol behaviour equation is replaced by three first order differential equations for the moments of particle size distribution by assuming a well stirred volume and a lognormal distribution and by neglecting the exponential term in the Cunningham correction. Additional first order simultaneous differential equations are included to generate desired output. Multiple control volumes are analysed sequentially.

Input:

- mechanism, model and program control options
- initial aerosol source distribution
- aerosol source rates and distributions and vapour source rate
- aerosol and vapour material properties
- control volume structural characteristics
- control volume atmospheric properties
- vapour and aerosol model parameters

The following parameters may be tables vs time:

- aerosol source rate and distribution
- vapour source rate
- aerosol source particle material density
- control volume and surface areas
- control volume atmospheric temperature, pressure, molecular weight and gas law constant
- turbulent energy dissipation rate
- control volume wall temperature and diffusive plating boundary layer thickness
- leak rate

Atmospheric viscosity may be a table vs temperature.

Calculation of coefficients:

The coefficients are calculated at each time step, the time dependence of varying properties being given by one of three interpolations specified by input.

Solution of the equations:

The equations are solved by a semi-stiff two point predictor-corrector method. The Runge-Kutta method is used for the initial and specified restart steps.

Output:

HAA-4A prints all the input data. Output data is printed with a frequency controlled by input and at specified times. The short-edit option contains total suspended mass and concentration, mass removed by each process and respirable-sized leaked mass; suspended particle size distribution characteristics; total removal rate, mass balance; and, for each component, the fraction of suspended mass, total respirable leaked mass, total source mass and mass leakage rate. The long edit adds values of all time independent input parameters and fractional rates of change of each moment of the size distribution by each process. Plots of time dependent input parameters and short and long edit outputs may be made. Total leak and fallout rates and particle size distribution parameters are saved on auxiliary files.

Modelling of physical processes:

HAA-4A calculates the following processes:

- three source components with independent time dependent distributions
- Brownian, gravitational and turbulent agglomeration
- particle growth by vapour deposition
- aerosol plateout due to gravitational settling
- Brownian diffusion to surfaces and thermophoretic deposition
- leakage, including path restriction by aerosol deposits

Aerosol models contain the following correction factors:

- Cunningham slip correction (without exponential term)
- Klyachko corrected settling velocity
- overall shape factor for mean particle size or constant
- separate constant dynamic and collision shape factors
- collision efficiencies for turbulent shear agglomeration and for gravitational and turbulent inertial agglomeration

The aerosol is assumed to be well stirred and to have a lognormal particle size distribution.

PWR-specific features:

The rate of vapour deposition is specified by input. The rate of growth of the particle is proportioned to the product of its area and the vapour flux to the particle surface, resulting in a rate of growth of surface independent of particle size.

Status of code:

The code has been verified against partial analytical solutions. Cases with suspended concentrations in excess of 10kg per cubic metre have been run. Comparison with dry aerosol experiments have been made up to concentrations of 200g per cubic metre have been made with good results. Shape factors, collision efficiencies and plating boundary layer thickness are chosen empirically.

Developments in progress include multiple vapour sources, multiple aerosol sources with time dependent material densities, calculation of aerosol deposition by diffusiophoresis and comparison with experiments containing steam. A forerunner HAA-3B has been compared to the CSTF test containing steam with encouraging results.

A 3.4 NAUA-4

H. Bunz et al, NAUA Mod 4, A code for calculating Aerosol Behaviour in LWR Core Melt Accidents, Kernforschungszentrum Karlsruhe, Report No. KFK-353, August 1983. This synopsis abstracted from Nuclear Aerosols in Reactor Safety, Nuclear Energy Agency, OECD, Paris 1985.

NAUA-4 is a computer code for calculating aerosol behaviour in closed volumes which contain a condensing steam atmosphere. The code was designed and developed for application to fission product transport and depletion during core melt accidents in LWRs especially PWRs. Main improvement over NAUA-3 was the substitution of the condensation model by a new experimentally verified one.

Programming strategy:

The integro-differential equation for aerosol behaviour in a closed container under stirred conditions is replaced by a set of coupled first order equations. The particle number density distribution function is replaced by a number of discrete monodisperse fractions. Size distributions are arbitrary, although for convenience in the program input, log normal functions may be used.

Input:

Input data are grouped in the following categories:

- control volume parameters
- aerosol process parameters
- numerical parameters
- program control parameters
- output control parameters
- aerosol source data
- time dependent thermodynamic properties
- time dependent leak rates
- restart and/or next compartment files

All this input is list directed or unformatted.

Calculation of coefficients:

Constant coefficients are calculated at the beginning of the program. Temperature and time dependent coefficients, aerosol source, steam source and leak rates are calculated every time step. Except for aerosol source and leak rate, all coefficients are dependent on temperature, which is input as a time function. The same is valid for the steam air ratio, assuming a fixed amount of air and a saturation steam concentration.

Solution of the equation:

The differential equation is solved by the Euler-Cauchy method. The code calculates the time step to ensure numerical stability. For the condensation process a separate routine using a separate (mostly smaller) time step is called.

Output:

Depending on the problem the following output can be chosen:

- short printed output
- long printed output
- restart data set
- leakage data set for next compartment
- plot data set for time functions
- plot data set for size distributions

Modelling of physical processes:

NAUA-4 calculates the following processes:

- Brownian agglomeration
- gravitational agglomeration*
- condensation/evaporation of steam on/from particles*
- gravitational settling
- Brownian diffusion
- leakage

(* can be activated by input parameters)

Diffusiophoresis was to be included by end of 1985.

Aerosol sources can be input via a three dimensional array of parameters containing 30 time intervals, 50 nuclide fractions and two size distribution modes. Each of the 3000 source modes may be instantaneous or at constant rate and with different effective density.

The concept of effective density is based on the spherification of particles by the condensation process. The option of using shape factors is still in the code to bypass the spherification for dry aerosol problems.

The aerosol leakage, including size distributions at every time step, can be written on a separate file for a sequential compartment calculation. The next compartment can be calculated in the same job.

A restart option has been integrated for saving all current data at the end of a calculation.

PWR specific features:

NAUA-4 has been developed for PWRs and is thus completely PWR specific. Moreover it is specific for meltdown accidents involving large masses of aerosols.

Status of the code:

NAUA-4 has been made available. Existing documentation is being updated and published (1985). Numerical methods have been checked by various comparison calculations. The single processes are experimentally verified with the exception of diffusiophoresis which is still on going. Agreement with NSPP 400 experiments is good when diffusiophoresis is included. The code was tested in DEMONA.

A 3.5 RESULTS OF THE GREST CODE COMPARISON EXERCISE

Abstracted from CSNI Report No 116 by H.A. Morewitz, F.J. Rahn and R. Sher, Nuclear Energy Agency, OECD, Paris, 1986

The GREST code comparison exercise was a major test of a number of aerosol behaviour computer codes applied to specified accident conditions in LWRs. The accidents modelled were:

1. AB hot leg sequence in the US Surry 1 PWR. This sequence was a LOCA resulting from a large break in the primary system hot leg combined with the failure of AC power to the engineered safety features. Molten corium/concrete interactions following reactor vessel melt through were included but ex-vessel steam explosions were not.
2. S2CD cold leg sequence in US Surry 1 PWR. This was a LOCA due to a small break in the primary system cold leg, combined with the failure of the emergency core cooling injection system and containment spray system, but with the containment safety systems in operation. Extensive corium/concrete reactions and ex-vessel steam explosions not considered.
3. Large hot leg break LOCA for the German Biblis B PWR. This assumes reactor cavity initially dry and corium/concrete interaction immediately on pressure vessel melt through; no containment sprays; ECCS operates but conversion to recirculation core cooling fails, leading to core melting.

Codes tested:

Code	Country	representation of aerosol size distribution
AEROSIM-M	UK	FINITE ELEMENTS
AEROSOLS/B1	FRANCE	"
SWNAUA	USA	"
NAUA Mod 4	USA	"
NAUA Mod 5	FRG	"
HAA-4	USA	MOMENTS METHOD
RETAIN 2C	FINLAND	"
RETAIN-S	SWEDEN	"
CONTAIN	USA	FINITE ELEMENTS
REMOVAL	JAPAN	"
HAARM-S	SPAIN	MOMENTS METHOD

All shape factors 1, density correction factor 1
size distribution mm radius micrometre sigma

Surry AB	0.5	2
Surry S2CD	1.5	2
Biblis LOCA	0.16	1.5

Containment:

Surry	volume 50,970 c metre	Biblis	72,000 c metre
	wall area 21,900 sq m		50,000 sq m
	floor 1,277 sq m		4,500 sq m
	leak rate 1% vol/day		2.5% vol/day

RESULTS:

1. There were appreciable differences in the results for most quantities of interest in all three problems. Differences were appreciable between results from different codes, from the same code used by different organisations, and from different versions of particular codes of the same type even when run by the same organisation.

2. The moments methods codes generally gave higher suspended mass concentrations and higher leaked mass than the discrete (finite element) codes.

3. All codes generated similar (generally to 1%) values of the total retained mass (plated plus settled mass), but the partition of the retained mass between plated and settled fractions showed large variations (presumably due to different condensation and diffusiophoresis models used).

4. All of the codes preserve (or force) mass balance. On the other hand even the small or residual mass unbalances are comparable to or larger than the total leaked mass.

5. Wet/dry effects vary considerably between the codes. In general HAA-4 showed the largest effects due to condensation on aerosol particles whereas NAUA-4 showed modest or negligible effects depending on whether EPRI or SWEC was using it. AEROSIM-2 showed small effects for SURRY AB but larger differences for the other two problems.

6. The differences due to using the Fuchs (F) or Pruppacher and Klett (P-K) gravitational agglomeration coefficients were generally less than a factor 2 in the integrated, leaked, plated and settled masses.

7. In general the Biblis problem resulted in more scatter among the results than the SURRY problems.

SUMMARY:

Surry AB:

Time integrated leaked mass: Dry aerosols spread of factor 3.6 for $t > 20,000$ s. Factor of about 1.6 due to F vs P-K. P-K higher;

Wet aerosols spread in leaked mass factor 2 at long times. 20% of spread accounted for by F or P-K

Total suspended mass: At 1000s 50% spread between results. At 173,000s difference between using F and P-K in RETAIN-2C was a factor 7. If RETAIN-2C is omitted, spread between remaining results, a factor 5. This appears due to whether the moments method or discrete value used in code, the latter giving lower results.

Suspended mass of CsOH: Results spread 2.5 orders of magnitude at 10,000s and 4 om at 100,000s. Lower limit with CONTAIN and AEROSIM-M, upper limit by HAARM-S. If these three are omitted, spread is a factor 8 at 100,000 s. The upper limit is then RETAIN-S and the lower NAUA-5, remainder are within a factor 2.

Suspended mass of CsI: Results spread over 3 om at 10,000s and 5 om at 100,000s. Upper limit HAARM-S and lower limit CONTAIN and AEROSIM-M. If these are omitted spread is factor 4 at 100,000s. Upper limit then HAA-4 and lower NAUA-5

Time integrated plated mass: If HAARM-S and calculation without Stefan flow (HAA-4 and NAUA-4) are ignored, spread in long term is a factor 4.5 between upper REMOVAL curve and lower AEROSIM-M curve. Use of F or P-K caused little difference, nor did water vapour condensation

Time integrated settled mass: For dry cases spread is 30% and for wet 80%

Mass median radius: for dry aerosols mmr ranged from 0.836 micrometres (RETAIN-C,P-K) to 1.75 (NAUA-5,P-K) at 1000s and from 0.91 (NAUA4,P-K) to 1.72 micrometres (RETAIN-S) at 86,400s. After 10,000 s the radii calculated by the moments method codes cluster about a value of 2um, the results of the discrete codes cluster about 1.5um. The results of the moments methods code show a peak size at about 40,000s which is absent in the discrete codes.

SURRY S2CD:

Time integrated leaked mass: For dry aerosols at 86,400s results range from 1322g (REMOVAL) to 3690g (RETAIN-S) a factor of 2.8. For wet aerosols at 86,400 range is from 1342g (HAA-4) to 1883g (NAUA-5) a factor of 1.4.

Moments method calculations give consistently higher (30-50%) than the discrete codes.

Total suspended mass: For dry aerosols the mass concentrations ranged from 25.5g/cubic metre (REMOVAL) to 36.8 g/cubic metre (RETAIN-S) at 1000s and from 0.061g/cubic metre (REMOVAL) to 0.6g/cubic metre (RETAIN-S) at 86,400s. Wet aerosol mass concentrations were from 14.2g/cubic metre (AEROSOLS/B1) to 2.1g/cubic metre (NAUA-5) at 1000s and from 0.274g/cubic metre (HAA-4) to 0.88g/cubic metre (AEROSOLS/B1)

Time Integrated Plated Mass: At 1000s the spread in results was a factor 7.3

Time Integrated Settled Mass: Plated plus settled mass (i.e. retained mass) relatively insensitive to presence of water vapour

Mass median radius: At 1000s the spread was a factor 9.6; at 10000s, 7.55 and at 86,400s 2.13. Usually 1.5 - 3.0 micrometres

BIBLIS:

Time integrated leaked mass: Spread in results a factor 8.5 RETAIN-2C (P-K) gave the high value and AEROSIM-M the low. Discrete codes give lower leaked mass than moments by roughly a factor 2. P-K in RETAIN-2C increases leaked mass by 50% over F.AEROSIM-M and HAA-4 give a factor of 2 to 2.5 between dry and wet (dry>wet)

Total suspended mass concentration: spread a factor of 2.5 among the discrete codes and 1.8 among the moments codes at 5,000s. From then on the moments codes give a higher value than the discrete codes. moments method curves tend to cluster more closely. At 86,400s the moments methods codes give values between 0.01 and 0.03 g/cm except for NAUA-4 which is four orders of magnitude lower.

Time integrated plated mass: More than an order of magnitude difference

Time integrated settled mass: If NAUA-5 is excluded range is about a factor 3. Wet/dry effects small ranging from negligible (NAUA-4) to about 20% (HAA-4)

Mass median radius: values cluster within a factor 2 up to 10,000s; at later times NAUA-4 shows lower values by a factor 3.

After about 2000s the mmr is about 1 micrometre.

A 3.6 CEC SENSITIVITY STUDY OF AEROSOL CODES IN LWR CONTAINMENT
by V.E. Della Loggia, CEC, Brussels, abstracted from IAEA-SM-281/39

The reference case for the study was the S2CD sequence of the SURRY 1 PWR. A small 2" diam break is assumed to occur in the cold leg of the primary, no steam explosion is assumed but the molten corium/concrete interaction that follows pv melt through is included. Thermal-hydraulic data deduced from results obtained by French code JERICO. Codes compared were NAUA-5, AEROSIM-M, AEROLOLS-B1 and CORRAL-2. Results compared were: suspended aerosol concentration, aerodynamic mass median radius and standard deviation of suspended particle distribution, settled mass on floor, diffused mass on walls, leaked mass and particle size distribution at specified times. The leaked mass for dry and wet cases calculated by the mechanistic codes are in reasonable agreement while CORRAL-2 (used in WASH-1400) gives conservative values in the long term (beyond 1000-10000s).

A 3.7 BEHAVIOUR AND RETENTION OF FISSION PRODUCTS IN THE CONTAINMENT

by W.O.Schock, W.Schikarski, F. Abbey and I.H.Dunbar
IAEA-SM-281/30, 1986

Abstract: Fission product retention in the containment is a key issue in estimating radiological source terms from accidents in LWRs. Since the publication of the Reactor Safety Study considerable progress has been made in many countries in the development of models and codes to describe fission product behaviour. The codes have been compared. Experimental investigations, both basic research and large-scale integral simulations were done and are being continued. The current status is that codes are believed to be almost in their final stage for application to source term assessment. The questions that remain include coupling to thermal hydraulics and the influence of spatial inhomogeneities in complicated containment geometries. Such influences, although believed to be of lesser importance, remain a possibility to alter source terms; their magnitude has still to be quantified. The results of calculations with the new codes show a reduction of the source term by orders of magnitude compared with those from the early risk studies.

The paper gives a good description of the development of aerosol codes and the significant features of some of them. In the Reactor Safety Study, modelling of aerosols was carried out using the CORRAL code which assumed a single settling rate for the aerosols which was a universal function of the time since release. Since the settling rate is a function of the aerosol source and the thermal-hydraulic conditions (the rates were based on the Containment System Experiments, which were not representative of reactor accidents), the CORRAL aerosol models have only limited relevance to accident modelling.

Mechanistic codes modelling aerosols were first developed under dry conditions for studies of sodium fires in severe fast reactor accidents. They can be divided into discrete codes which divide the aerosol size range into discrete size classes, and log normal codes which make the more restrictive assumption that the size distribution is always log normal. In comparisons it has been found out that for dry conditions the discrete codes are in good agreement and the one log normal code tested diverged significantly from the discrete code. The discrete codes were also in better agreement with the experimental results.

The first aerosol behaviour code to incorporate steam condensation on to particles was NAUA developed in the FRG. This code is now widely used in the USA, Sweden, Finland, Denmark, Italy and Spain. More recently, following experiments, it has had a diffusiphoresis model added to it. In the last few years two European fast reactor codes have been extended to take account of the effects of condensing steam - AEROSIM-M in the UK

and AEROSOLS/B1 in France. In the USA similar developments led to the codes HAA-4, QUICK-M and the MAEROS model in CONTAIN, and in Japan, to the REMOVAL code.

The major difference between the codes is their modelling of condensation on the particles. They have different solutions to the problem of where to make the boundary between the aerosol code and the thermal-hydraulic code from which it takes its input. AEROSIM-M takes as its input the actual mass of water predicted by the thermal-hydraulic code to be condensed on to the aerosol. The only part of the modelling of the condensation used is the dependence of relative condensation rates on particle size.

NAUA takes the same water mass as AEROSIM-M. It then calculates steam supersaturation and then the condensation rate on to the particles depending on particle size, taking into account the curvature of the particle surface. Care has to be taken with this approach as the condensation rate calculated by NAUA has to be nearly the same as that predicted by the thermal-hydraulic code from which other input is taken. Otherwise the difference in the release of latent heat would make the two calculations inconsistent. If there are enough particles present the condensation will be fast compared with the rate of change of the thermal-hydraulic properties and equilibrium will be restored in each time step, as is assumed in the thermal-hydraulic code. The mass of water condensed will then equal the input water mass.

AEROSOLS/B1 requires the supersaturation ratio as its input, from which it calculates condensation on soluble particles only, including the curvature effect. As with NAUA, consistency with the thermal-hydraulic data has to be maintained.

Once condensation has been modelled one has to distinguish between the water and the non-water mass in the calculation. It is also useful to be able to follow separately radiologically important nuclides. The question then is whether to allow the aerosol composition to be a function of particle size or to leave it independent. NAUA distinguishes between water and non-water mass as a function of size and can follow up to 50 nuclides on a size independent basis. AEROSIM-M has size dependent modelling of up to ten components, one of which can be water. MAEROS similarly uses eight components.

Subject to experimental validation and with the exception of the modelling of the condensation of water, the stand-alone aerosol codes are virtually in their final stage. There is still scope for adding on further facilities and improving the details of the physics, but it is unlikely that the overall trend of the predictions will be altered. On the other hand scientifically correct modelling of steam condensation requires a coupled thermal-hydraulic and aerosol code. Therefore no stand-alone code is an exact solution to the condensation problem. However existing stand-alone aerosol codes MAY be adequate for the

purposes of reactor safety analysis.

In the GREST comparison the log normal codes give consistently higher values of the accumulated leaked mass (the source term). Within a code group the scatter in the predicted leaked masses is only a factor of two.

EFFECT OF BOUNDARY CONDITIONS

To calculate fission product in the containment one needs details of timing and nature of fission product release, thermodynamic details inside the containment (temperature, pressure, gas composition and condensation rates) as functions of time. Also need details of plant geometry - the surface areas, however difficult to estimate, are sensitive input for removal mechanisms. Stand alone thermal hydraulic codes ignore droplet curvature and solute effects and put the condensed water into droplets of a uniform settling rate. The CONTAIN code is a fully coupled thermal hydraulic and multicomponent aerosol code. Both sorts of processes interact at each time step. At present it does not contain the effects of curvature and solutes on the condensation, but is a framework into which these could be incorporated. (Bergeron et al, 1985).

EXPERIMENTAL

The behaviour of aerosols in LWR accidents is being investigated experimentally in NSPP at Oak Ridge. Facility is a 38m thermally insulated SS vessel, heated to operating conditions by steam injection, aerosols injected by vapourising powders (including uranium) by plasma gun. Single component tests with concrete, U_3O_8 , Fe_2O_3 and showed different removal rates - concrete aerosol removed more slowly than the others and did not depend on steam being present. Others were removed more rapidly in condensing conditions. Differences explicable by morphology of particles with density shape and amount of steam condensing. The first test in a condensing atmosphere was compared with NAUA calculations and there were large discrepancies until diffusiophoresis was included in the NAUA code.

DEMONA

DEMONA models a core melt accident in a quarter scale model of the BIBLIS A station which can be operated at the correct temperature and pressure. Aerosols are produced by vapourising and oxidising powders of iron, silver and tin and concentrations of over 10g/cubic metre can be achieved. Experiment simulates a late overpressure failure sequence in a German large dry PWR.

Results of wet tests can be summarised:

Condensation of steam on particles leads to much faster removal of the aerosol. In the dry tests the aerosol concentration fell below the limit of detection after 48 hours, this condition was reached in the wet tests in 8 to 12 h. Onset of steam

condensation could be directly observed by two independent measurements and the time was in good agreement with predictions from the thermodynamics code. Comparison of measured and computed concentrations (NAUA) was good. This is in spite of the fact that the DEMONA building is a multi-compartment one and NAUA is a single compartment code. One of the aims of DEMONA is to see whether a single compartment code can model effects in a structured building with sufficient accuracy.

Conclusion from both NSPP and DEMONA is that no deficiencies in the NAUA code have been found.

FILTERED VENTING

Retention of fission products in the containment very effective as long as integrity is preserved. At Barseback (Sweden) a 10,000 cubic metre gravel bed has been installed. Results of experimental and analytical work showed that for 24 hours complete condensation of the steam was obtained and a retention of > 99.9% of particles and elemental iodine established.

SUMMARY

Schock says that "recent findings support the earlier hypothesis that the predominant form of iodine entering the containment is CAESIUM IODIDE AEROSOL and that the remaining caesium is in the form of caesium hydroxide aerosol". (no supporting references given). Use of improved models and codes has led to significant reduction in source term values. e.g. use of NAUA to the large loss-of-coolant-accident initiated core melt accident with overpressure failure of the containment (delta failure mode in WASH 1400 terminology) reduces the fraction of iodine released to the environment from 0.7 to 0.0004, caesium from 0.5 to 0.0004 and tellurium from 0.3 to 0.002. It is believed that the error in the leaked mass from the containment is less than a factor five.

CONCLUSIONS

Retention of fission products in the containment under severe accident conditions is predominantly influenced by the nuclear aerosol behaviour. Modelling of aerosol behaviour in today's aerosol codes is well advanced and large scale validation experiments are underway. It is considered that the calculational uncertainties in aerosol codes are probably small compared with those in other parts of the source term calculation.

A 3.8 AEROSOL MODELLING IN CONTAINMENT

by H. Jordan and V. Kogan abstracted from IAIA-SM-281/45, 1986

The degree to which particle growth by condensation and Stefan flow to containment surfaces accelerates removal of aerosol particles is governed by the thermodynamics of the containment system coupled with the behaviour of the aerosol. Some preliminary results of calculations with the QUICKM code designed to explore this coupled thermodynamic/aerosol behaviour in as mechanistic a way as possible are presented. Calculations performed on a Surry TLMB sequence and show unexpected sensitivity to the numerical treatment of the coupled models and to the degree of coupling. The results are also sensitive to assumptions on the transition of two phase flow from the pressurised primary system to the containment. BECAUSE THE SATURATION RATIO IS A SENSITIVE FUNCTION OF GAS TEMPERATURE, ITS PREDICTION IS PARTICULARLY UNCERTAIN. SINCE THE SATURATION RATIO DRIVES STEAM CONDENSATION ON TO PARTICLES THE CONCLUSION IS THAT FURTHER WORK IS NEEDED BEFORE ANY PREDICTION OF WATER VAPOUR CONDENSATION ON PARTICLES IS BELIEVABLE.

Points out that the two codes NAUA and CONTAIN are widely used in assessing aerosol behaviour under wet LWR containment conditions. NAUA is a single aerosol component code (which does track composition with time) which relies on other codes to present it with airborne water vapour available for condensation on particles. It cannot therefore model possible competition between particles and wall surfaces for condensing steam. Thus there is no feedback to containment atmosphere thermodynamics from aerosol phenomena in the NAUA treatment. It is conceivable however that condensation on particles can dominate the vapour/liquid transition in containment. Aerosol loadings as low as $1\text{g}/\text{m}^3$ present more dispersed surface area than wall surfaces and mass transfer coefficients to aerosol particles are orders of magnitude higher than those for walls. If supersaturation can be obtained, driving forces for condensation on to walls and on to particles are of the same order.

The CONTAIN code, while it does couple condensation on particles with the thermodynamics of the containment, does so over an arbitrary user-determined time step. The adequacy of this approach needs verification. The reason is that condensation on particles is an extremely rapid process and proper partitioning of condensate between walls and particles would require simultaneous solution of all the differential equations involved, or, if these are solved sequentially (as in CONTAIN), then over a time step that is of the order of the time constant for condensation on particles. This time step is far too small for the CONTAIN code.